FEUP FACULDADE DE ENGENHARIA
UNIVERSIDADE DO PORTO

INTEGRATED MASTER IN ENVIRONMENTAL ENGINEERING

2013/2014

Indoor Ultrafine Particles: Evaluation of Home Environments

Cátia Raquel dos Santos Teixeira

Dissertation submitted to obtain the degree of

MASTER IN ENVIRONMENTAL ENGINEERING

President of the Jury: Cidália Maria de Sousa Botelho

Auxiliary Professor of the Department of Chemical Engineering of the Faculty of Engineering of University of Porto

---

Supervisor: Maria do Carmo da Silva Pereira

Auxiliary Professor of the Department of Chemical Engineering of the Faculty of Engineering of University of Porto

Co-supervisor: Klára Slezáková

Post-Doctoral Researcher

Porto, July 2014

Faculty of Engineering University of Porto

Indoor Ultrafine Particles: Evaluation of Home Environments

Cátia Raquel dos Santos Teixeira

Dissertation for the Degree of

Integrated Master in Environmental Engineering

Supervisor: Maria do Carmo da Silva Pereira

Co-Supervisor: Klára Slezáková

Porto, July 2014

Abstract

Ultrafine particles (UFP) are defined as particles with aerodynamic smaller than 100 nm. Particles this small are of great concern in terms of public health, since they are able to penetrate deep in the respiratory system and reaching the alveolar wall and bloodstream, depositing on specific organ tissues. The complexity of indoor UFP exposure (spatial variability, indoor sources, infiltration of UPF from various outdoor emission sources, seasonal variability in concentrations and composition) indicates the need to further study this pollutant in order to fully comprehend its impacts on human health. This is especially relevant for sensitive group such as children. Considering the fact that children spend a great amount of their time in homes, an analysis of these environments regarding UFP is worthy of note. Thus this work aims to evaluate UFP home environments. The specific objectives were: (i) to assess ultrafine particle number concentration in four Portuguese homes; (ii) to evaluate potential emission sources of ultrafine particles in home environments; and (iii) to estimate exposure dose of 3 to 5 years old children to ultrafine particles in home environments.

UFP particle number concentrations were sampled in four Portuguese urban and rural homes (one with smokers and three non-smokers) during a total of 38 days. UFP were sampled concurrently both indoors and outdoors (between 00:00 to 23:59). In addition, different fractions of particulate matter (namely PM_{10}, $PM_{2.5}$, and PM_1) as well as, temperature and relative humidity were also collected in order to better characterize the indoor environments.

The results showed that the average indoor UFP number concentration levels were dependent on the type of existing sources, but were generally higher if observed the following factors: (i) smoking occupants and (ii) small room volume. UFP number concentration obtained in homes with where the occupants were smokers has the highest level of all homes (1.64×10^4 particle cm^{-3}). Non-smoking homes UFP levels showed a linear reverse dependence on room volume (1.09×10^4; 1.11×10^4 and 1.24×10^4 particle cm^{-3}, in order of decreasing room volume). Regarding outdoor ambient concentration levels of UFP, the higher UFP mean number concentration were observed in urban areas (8.05×10^3; 1.05×10^4 and 1.24×10^4 particle cm^{-3}) in opposition to rural location (7.78×10^3 particle cm^{-3}). I/O ratios were bigger than 1 in all homes, indicating that emissions from indoor sources were the main contributor to indoor UFP levels. Higher exposure doses of UFP yield the result that smokers home presented higher values than non-smokers ones. Indoor PM values were also higher in the home of smokers for all fractions (1.34×10^2; 1.36×10^2 and 1.30×10^2 µg m^{-3} for PM_1, $PM_{2.5}$ and PM_{10} respectively). Concentrations obtained for PM_1 in non-smokers residences were 1.81×10^1; 2.86×10^1 and 2.59×10^1 µg m^{-3}. $PM_{2.5}$ resulted in concentrations of 1.93×10^1; 2.99×10^1 and 4.76×10^1 µg m^{-3} with non-smoking occupants while PM_{10} levels were 2.16×10^1; 3.27×10^1 and 5.13×10^1 µg m^{-3}. Outdoor $PM_{2.5}$ was also measured, allowing calculating I/O ratio for this fraction. The results obtained for I/O ratios were bigger than 1 for three homes (including the home of smokers), indicating than indoor sources of $PM_{2.5}$ were the main contributor for indoor concentration levels for this particle fraction; only one urban home exhibited outdoor $PM_{2.5}$ as the main contributor for indoor concentrations of this size range (with I/O lower than 1).

Evaluation of indoor UFP producing sources resulted in the conclusion that cooking activities, specifically usage of electric oven, usage of electric toaster and boiling are the activities that produce higher number concentration levels of UFP. Smoking resulted also in a major increase of indoor UFP number concentration but also to the perpetuation of these levels longer in time compared with other sources and acting as a precursor for PM formation as observed in the obtained results.

"A human being is a part of the whole, called by us the «Universe», a part limited in time and space. He experiences himself, his thoughts and feelings as something separated from the rest, a kind of optical delusion of his consciousness. This delusion is a kind of prison for us, restricting us to our personal desires and to affection for a few persons nearest to us.

Our task must be to free ourselves from this prison by widening our circle of compassion to embrace all living creatures and the whole of nature in its beauty."

Albert Einstein

in H. Eves - Mathematical Circles Adieu (Boston, 1977)

Acknowledgements

My following words are dedicated to all of those who helped making this work possible and to whom I owe my gratitude for that achievement.

I would like to thank my supervisor and co-supervisor, Professor Maria do Carmo da Silva Pereira and Klára Slezáková for helping and guiding on the development of this work. Thank for all your precious teachings, suggestions and advices, patience and understanding and for your special contribution to both academic and personal growth.

This work would not be possible without the cooperation and good will of the owners of the sampled homes, who patiently tolerated campaign weeks inside their residences. Not specifying names in order to maintain their privacy, I would like to dedicate my word of appreciation to all the inhabitants that live in each of the four homes studied in this work.

To my family, words cannot describe my deepest and largest gratitude for all the opportunities I was given by them. To my parents, Manuel and Fátima Teixeira who always supported me on the entire academic and life path, providing and encouraging me to follow my goals, and teaching me the difference between cost and value. I owe them for raising me to be an honest, determined woman, who is curious about all fields of knowledge. To my brother, Ricardo Teixeira, for the influence, support and example given over my entire life, to whom I owe my interest in Engineering, to begin with.

I would also like to mention the family of "Minas e Ambiente" that I witnessed to grow not only in number, but also in the personal development of each individual. My respect to all of you the ones that taught and learned, the ones that teach and learn and the ones that learn and will teach, keep up the good work, excel yourselves, ourselves. To 08, above and bellow, "thank you would be what I would say, if I could".

I would also like to thank to the Faculty of Engineering of the University of Porto, to all the teachers and students for directly or indirectly making me an Engineer, both on academic and personal level.

I dedicate this work to João Quintanilha.

This work was supported by the project PTDC/DTP-SAP/1522/2012|FCOMP-01-0124-FEDER-028797.

List of Publications

Most of the content in this thesis was submitted and accepted for publication in:

Publications in Scientific Meetings with abstract:

K. Slezakova, C. Teixeira, J. Fonseca, S. Morais, M. C. Pereira, Exposure of 3-5-years old children to indoor ultrafine particles: assessment of homes and schools, *3rd International Congress on Environmental Health*, Porto September 2014.

Table of Contents

List of Figures

List of Tables

Nomenclature

Abbreviations

A	Auto-estrada
APA	Agência Portuguesa do Ambiente
BMDR	"Black Magic Dust" Residences
CO	Carbon Monoxide
CO_2	Carbon Dioxide
COPD	Chronic Obstructive Pulmonary Disease
D	Dose Rate
EU	European Union
H	Home
I/O	Indoor/Outdoor Ratio
NO_X	Nitrogen Oxide
NR	Normal Residences
O_3	Ozone
PM	Particulate Matter
PM_1	Particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 1 µm aerodynamic diameter
$PM_{1-2.5}$	Particulate matter fraction particle size ranged from 1µm to 2.5 µm

$PM_{2.5}$	Particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 2.5 µm aerodynamic diameter
PM_{10}	Particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter
$PM_{2.5-10}$	Particulate matter fraction particle size ranged from 2.5µm to 10 µm
RS	Rural School
SO_X	Sulphur Oxide
US	Urban School
U. S. EPA	United States Environmental Protection
UFP	Ultrafine Particles
VCI	Via de Cintura Interna
VOC's	Volatile Organic Compounds
VRI	Via Regional Interior

Greek Symbols

∑	Sum

Roman Symbols

BR	Breathing Rates
BW	Body Weight
C	Particle number concentration

D_{ae}	Aerodynamic Diameter
N	Daily total time spent by age-specific children in home
n	Number of hours spent by age-specific children in that location per day
SD	Standard Deviation

Subscripts

i	Counter
WA	Weighted Average

1. Introduction

1.1 Relevance and Motivation

Air pollution is one of the main reasons for a high number of premature deaths and increased morbidity rates in the world. According to World Health Organization (WHO), in 2012 seven million deaths were caused by air pollution (WHO, 2014). Particulate matter (PM) is one of the main constituents of air pollution and may cause a vast number of negative consequences. A number of adverse effects on public health, especially on children and the elderly may arise in the exposure to this air pollutant. Respiratory tract complications like bronchitis, pneumonia and allergic reactions are some of the consequences associated with PM (Raaschou-Nielsen et al., 2010). It has become of special importance the study of PM implications on public health.

Ultrafine particles (UFP) relates to the smallest fraction of PM. smaller than 100 nm and less are then able to enter bloodstream, penetrating cell membranes and depositing on secondary and vital organs (Burtscher and Schuepp, 2012). With a relative high surface area compared to its volume, UFP may act as a transportation media into organisms and carry adsorbed toxic substances. UFP are present in the ambient environment, typically resulting from the emission of combustion gases, with motor engines being the predominant source in the urban environments (Morawska et al., 2008). Indoor sources of UFP mainly include cooking activities like frying, grilling, boiling, toasting and the usage of oven (Zhang et al., 2014). UFP number

concentrations may reach extremely high levels indoors compared to outdoor ones, since the confined space prevents the natural dispersion of this pollutant for a longer period of a time.

The exposure to indoor UFP is complex and its various issues such as spatial variability, indoor sources, infiltration from outdoors, and variability in concentrations have yet to be addressed. Furthermore, in a view of the amount of time that people spend indoors, the characterization and analysis of indoor UFP is of high relevance and urgency in order to improve and protect the well-being of both children and adults.

The aim of this study is to obtain information of PM focusing indoor ultrafine number concentration levels in home environments. These results may then allow further understanding on the behavior and dynamics of this air pollutant in indoor environments, more specifically in homes. The assessment of the potential risks of exposure for children living in these environments is also a relevant motivation for the execution of this work; epidemiological studies have showed that the exposure to UFP in a young age may be determinant on the good and healthy development of children and to their well-being later in life (Bernstein et al., 2008)

1.2 Objectives

This work aims to evaluate ultrafine particle in home environments located in the north region of Portugal. The specific objectives of this work were:

- To assess ultrafine particle number concentrations in four Portuguese homes in comparison with other studies;
- To evaluate indoor emission sources and the effect on ultrafine particle number concentration as well as potential outdoor sources;
- To estimate exposure dose of 3 to 6 years old children to ultrafine particles in their home environments.

1.3 Thesis Outline

This thesis is divided in 5 chapters, each associated with the following content.

Chapter 1, the present chapter, presents the motivation for the realization of this work as well as structure and outline of the thesis.

Chapter 2 refers to the actual state of the art, introducing the theoretical concepts regarding air pollution as well as the problematic associated with the subject. Focusing on ultrafine particles and particulate matter, it describes its properties, sources, formation processes, composition and environmental and health effects. A brief description of the applicable legislation as well as exposure assessment is also present in this chapter.

Chapter 3 describes materials and methods applied in the development of this work. A characterization of the studied residences, equipment characteristics and procedure of handling as well as a description of data treatment and statistical analyses is described in the chapter in question.

Chapter 4 consists of the obtained results and their discussions. It contains analysis of obtained indoor and outdoor concentrations and comparison with international studies, evaluation of particle sources, discussion concerning particle daily profiles for the different home environments, and exposure assessment.

Chapter 5 indicates future research needs and the conclusions reached through the execution of the present study.

1.4 References

BERNSTEIN, J. A., ALEXIS, N., BACCHUS, H., BERNSTEIN, I. L., FRITZ, P., HORNER, E., LI, N., MASON, S., NEL, A., OULLETTE, J., REIJULA, K., REPONEN, T., SELTZER, J., SMITH, A. & TARLO, S. M. 2008. The health effects of nonindustrial indoor air pollution. *Journal of Allergy and Clinical Immunology,* 121**,** 585-591.

BURTSCHER, H. & SCHUEPP, K. 2012. The occurrence of ultrafine particles in the specific environment of children. *Paediatric Respiratory Reviews,* 13**,** 89-94.

MORAWSKA, L., RISTOVSKI, Z., JAYARATNE, E., KEOGH, D. U. & LING, X. 2008. Ambient nano and ultrafine particles from motor vehicle emissions: characteristics, ambient processing and implications on human exposure. *Atmospheric Environment,* 42**,** 8113-8138.

RAASCHOU-NIELSEN, O., HERMANSEN, M. N., LOLAND, L., BUCHVALD, F., PIPPER, C. B., SORENSEN, M., LOFT, S. & BISGAARD, H. 2010. Long-term exposure to indoor air pollution and wheezing symptoms in infants. *Indoor Air,* 20**,** 159-167.

WHO 2014. Burden of disease from Household Air Pollution 2012 - Summary of results. Geneva: World Health Organization.

ZHANG, Q., AVALOS, J. & ZHU, Y. 2014. Fine and ultrafine particle emissions from microwave popcorn. *Indoor Air,* 24**,** 190-198.

2. State of the Art

2.1 Air Pollution

As we stand in a time where human kind is becoming progressively more conscious about the nature of what surrounds us, interrelations between all systems of environment have become evident. The adverse changes people have caused to a specific system or subsystem have been unavoidably affecting all existing environmental systems. The urge to take responsibility for these actions has become obvious, since human have been forced to deal with the consequences of irresponsible acts from the past (Ramanathan and Feng, 2009).

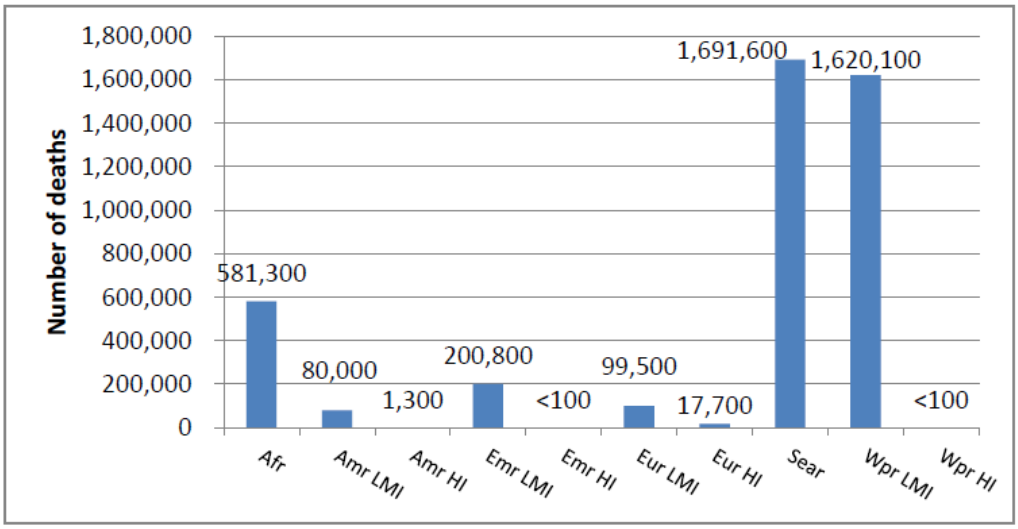
Air pollution has started as early as man discovered fire, but its global negative effects shown only in the 19^{th} and 20^{th} centuries when the need to produce goods in a large scale led to the development of fossil fuel combustion processes (Slezakova, 2009). The Industrial Revolution was the first big boost in the technological development, driving civilization through a franticly rapid growth rhythm, mainly concerned with satisfying demands and economic profit. Since fossil fuel combustion processes were boundlessly applied as the main source for energy production and transportation in addition to its industrial application, chemical compounds that were not part of the original atmosphere of the planet started to integrate the troposphere (Ramanathan and Feng, 2009). The lack of a specific legislation in the past regarding this phenomenon, as well as a generally reckless attitude about the consequences of emitting combustion gases directly into the atmosphere, brought air quality to a concerning degree of pollution escorting this unsustainable development through time. The most common pollutants

of an anthropogenic origin are volatile organic compounds (VOCs), ground-level ozone (O_3), carbon monoxide (CO), carbon dioxide (CO_2), nitrogen oxides (NO_X), toxic metals (such as mercury or lead), sulphur oxides (SO_X), particulate matter (PM) and persistent free radicals (that can be attached to PM).

Nowadays, even though human population is still extremely dependent on fossil fuel combustion processes to support every-day life, technology on every sort has also been progressing towards the mitigation of the unfavourable consequences of pollution upon the environment. This awareness evolved as the most severe outcomes were observed affecting all the other systems on earth because of this occurrence. Well-documented incidents have been reported about these repercussions. From the vastly known Great Smog in London in 1952, that caused the death of approximately 12,000 people, until the recent Paris pollution incident where several short-term effects on the city's residents have become world news (Alves and Mariano, 2014; Ribeiro, 2014) . Recent epidemiologic studies demonstrate the effects of air pollution in public health (Slezakova et al., 2013a). Short-term effects can cause allergic reactions, bronchitis and pneumonia as well as irritation of the upper respiratory system including eyes, nose and throat (Raaschou-Nielsen et al., 2010). Long-term effects can arise as lung cancer, heart disease or chronic respiratory disease (Bruce et al.; 2000; Mu et al.; 2013; Zhang et al., 2009). These effects are even more aggravated if children exposure and the elderly are considered. Ultimately, for the most acute exposures the outcome can end in an increase of mortality rates due to implications of resulting from cardiovascular and respiratory diseases (Viegi et al., 2004).

The quality of indoor air is affected by outdoor air pollution, allied to the fact that numerous sources of air pollution may exist indoors. Cooking, heating systems, the presence of pets and the type of furniture material are some of the factors that may produce particulate matter indoors (Spilak et al., 2014). Since air indoors is far more confined that outdoors, preventing the dispersion of pollutants, its concentration can reach concerning levels. Fourteen hours is approximate amount of time spent daily at home by a Portuguese family, thus revealing the importance of a healthy air inside their dwellings.

A recent report on air pollution published by the WHO shows preoccupying statistics about this matter. Seven million deaths were caused by air pollution in 2012 (WHO, 2014). This means that one in eight of total global deaths were a result of exposure to air pollution. Of these, 3.7 million deaths were attributed to ambient air pollution and 4.3 million deaths were due to household air pollution. These findings are twice higher than previous estimates and suggest that air pollution is nowadays the world's largest single environmental health risk (WHO, 2014). In addition, it reveals that indoor air pollution is the bigger contributor to air pollution-related deaths, underlining the urgency to its mitigation but also the need to further study these types of environments.



HAP: Household air pollution; Amr: America, Afr: Africa; Emr: Eastern Mediterranean, Sear: South-East Asia, Wpr: Western Pacific; LMI: Low- and middle-income; HI: High-income.

Figure 2.1.1- Total deaths attributable to household air pollution in 2012 by region (adapted from, WHO 2014).

2.2 Indoor Air Quality and Exposure

Indoor air quality can be defined not only by the presence of specific air pollutants but also by the level of comfort and perception that each individual makes of the air quality. In addition to the acceptable concentration levels of a number of substances, other factors like temperature, humidity and odours are conditions that contribute to indoor air quality (APA, 2014). Since nowadays people spend more than 80% of their time indoors, it is vital to understand exposure consequences and guarantee healthy air quality conditions in home environments (Hulin et al., 2010).

In general the environments can be classified into three different categories: rural or urban, indoor or outdoor and developing and developed countries (WHO, 2006). Rural environments often exhibit lower levels of air pollution in comparison to urban levels due to lower traffic density, less industrial activity and more distance between sources, allowing pollutants dilution in the environment resulting in lower levels both indoors and outdoors. Indoor levels are often higher than outdoor concentration because of indoor sources producing high pollution and the confined space. Inadequate ventilation can increase indoor levels of pollutants by not allowing enough outdoor air to dilute emissions from indoor sources and by not carrying indoor air pollutants out of a confined space of dwellings (Dimitroulopoulou, 2012). High temperature and humidity levels can also slightly increase concentrations of some pollutants. Therefore, indoor concentrations of hazardous aerosols can often be much higher than ambient air pollution levels (EPA, 2012). Developing countries show higher UFP and PM indoor levels than developed countries, overall because of the use of high polluting sources for energy and heating, lack of a specific legislation and low economical resources. Indoor pollution in developing countries is mainly influenced by the use of simple biomass stoves(Gall et al., 2013).

Indoor exposure to air pollution can be calculated by the concentration of pollutants inside people's dwelling and, most importantly, by the time each individual spends in the polluted environment (WHO, 2006). This concept allows determining the actual dose for each individual, quantifying the amount of air pollutant inside the organism. It can be affected by many factors such as the exposure via, lung region surface area or breathing pattern, the source location in relation to the occupant as well as indoor air distribution and air mixing and the duration of this exposure (Donghyun and Novoselac, 2010). It is important to understand that the actual pollutant concentration inhaled by an occupant can be significantly different from indoor room concentration due the non-uniform airflow around human body. Caused by the temperature gradient between the individual and indoor air, occupants' thermal plume can transport a pollutant around the body toward the breathing zone (Donghyun and Novoselac, 2010).

For indoor air quality, with special focus to particles, it is important to understand the two main factors that influence exposure: i) outdoor originated pollutants that penetrate indoor environments; and ii) indoor generated pollutants from specific emission sources. This means that indoor particles are a mixture of outdoor infiltrated particles, particles emitted from indoor

sources and particles formed indoor from gas-phase reactions originated from both indoor and outdoor (Morawska et al., 2013).

It is clear that outdoor air greatly alters the air quality inside buildings. In fact, in the absence of indoor sources, the concentrations of indoor pollutants are governed by natural and mechanical ventilation and infiltration from outdoors. The equilibrium between indoor and outdoor particle concentration is known as infiltration factor and it is influenced by three main variables: i) particle penetration efficiency, ii) deposition loss rate, and iii) air change rate (Spilak et al., 2014). These parameters are mainly governed by outdoor conditions and building characteristics. Particle penetration efficiency represents the fraction particles from outdoor that have entered into the indoor confined space by window opening, leakage paths, gaps and building cracks. Deposition loss relates to the rate of particle settling onto interior surfaces. This also depends on the physicochemical properties of particles, such as its mass, shape and reactiveness. Air exchange or ventilation rate has a big influence on indoor pollutants concentration. If the room is not often ventilated it can lead to accumulation of pollutants, resulting in a high concentration levels during an extended period. Otherwise, if air change rate is elevated, pollutants are easily dispersed into the atmosphere but outdoor pollution may influence negatively indoor air quality (Rim et al., 2013).

Direct and indirect methods can be applied for the assessment of indoor exposure. Direct measurements can include stationary passive or active sampling in the individual under study. Indirect methods include indoor source inventories and questionnaires also accounting the amount of time people spend indoors and environmental concentration data (Viegi et al., 2004).

While there is a considerable toxicological evidence of potential detrimental effects of UFP on human health, the existing body of epidemiological evidence is insufficient to reach a conclusion on the exposure-response relationship of UF particles (WHO, 2006).

2.3 Particulate Matter

Particulate matter is defined as the complex mixture of both solid and liquid phases of organic or inorganic particles suspended in the air, originated from the most extensive natural and anthropological sources (WHO 2011). Characterization of these particles can take into account its many variable properties, but the most common definition takes into account the particle's aerodynamic diameter (D_{ae}). It is defined as the diameter of a spherical particle that has the

same inertial properties and settling velocity in air as the particle in consideration (Slezakova et al., 2013b). Although particles are not generally spherical, knowing its equivalent aerodynamic diameter allows predicting its aerodynamic properties in a fluid, specifically air. This characteristic is commonly designated simply as "particle size". The comparison between particles of different densities and irregular shapes allows study of their transportation, deposition and removal processes. Since many pollutants may be adsorbed on PM surface, particle size is also an important parameter when estimating its surface area, hence determining the reactiveness of the particle with its surroundings (environmental and biological systems).

Optical diameter is used to calibrate optical particle sizing instruments, corresponding to the diameter of a spherical particle that has an identical refractive index as the particle. The Stokes diameter is defined as the diameter of a sphere with the same density and settling velocity as the particle. For particles greater that 0.5 μm, the aerodynamic diameter can be defined as the product of the Stokes particle diameter and the square root of the particle density (Boubel et al., 1994).

The aerodynamic diameter of PM ranges from 20 nm up to 100 μm. The smaller the aerodynamic diameter of the particle, the more adverse effects it might cause as it can penetrate deeper into different systems (Slezakova et al., 2013a).The size of a particle is determined by how the particle is formed. For example, combustion typically generates very small particles, whereas coarse particles have their origin in mechanical processes, such as erosion (Boubel et al., 1994). They can be emitted by natural or anthropological sources. Forest fires, volcano activity, sea sprays, pollen, bacterial and virus matter, erosion and suspension of soil and dust from roads are some examples of possible natural sources of PM. Anthropogenic sources are associated with combustion related activities, especially fossil fuel burning in industrial processes for producing energy and goods, but also in some commercial activities and in personal transportation and heating.

Many existing classification systems use the standardized aerodynamic diameter as a parameter, also aggregating groups of particles regarding other relevant factors, depending on characteristics the most useful for the study case. The most common systems used by the scientific community are (Slezakova et al., 2013b):

i) Sampler cut-point;
ii) Regulatory classification;

iii) Occupational classification;

iv) Modal classification.

The sampler cut-point system classifies a size range of particles considering selective size groups relative to their special importance in a determined objective of study (public health, environmental impact, sources, etc.). It refers to a collection of particles, bellow or within a specific aerodynamic size with a 50% collection efficiency. This means that in a sampler for $PM_{2.5}$, the upper 50% cut-point is located at 2,5 μm of aerodynamic diameter (Slezakova et al., 2013b).

The regulatory classification system is relative to legislation concerning the need to monitor these particles concentrations in the atmosphere, in order to ensure quality of human health. PM_{10} and $PM_{2.5}$ were selected as indicators. The relevance of this choice is explained by the fact that PM_{10} are small enough to enter the thoracic region and $PM_{2.5}$ standard was based on epidemiological studies (Slezakova et al., 2013b).

The occupational classification system categorizes particles taking into account how deep they are able to deposit in human respiratory system, classifying its levels of potential health risk. The levels are considered inhalable particles, thoracic and respirable particles. According to its size, the smaller they are, the deeper they can penetrate on the respiratory system. The larger-sized particle deposit on the upper respiratory system. Thoracic particles can get past the larynx, depositing on the lower respiratory tract. Respirable particles can deposit as far as the alveolar level. In terms of correspondence with the size cut-point system, thoracic particles are often used as a synonym for PM_{10}. As for respirable particles, they are frequently used as an equivalent for $PM_{2.5}$.

The modal classification is based upon particle size distributions, formation processes, composition, sources and deposition pathways. It divides PM into two main modes: coarse and fine.

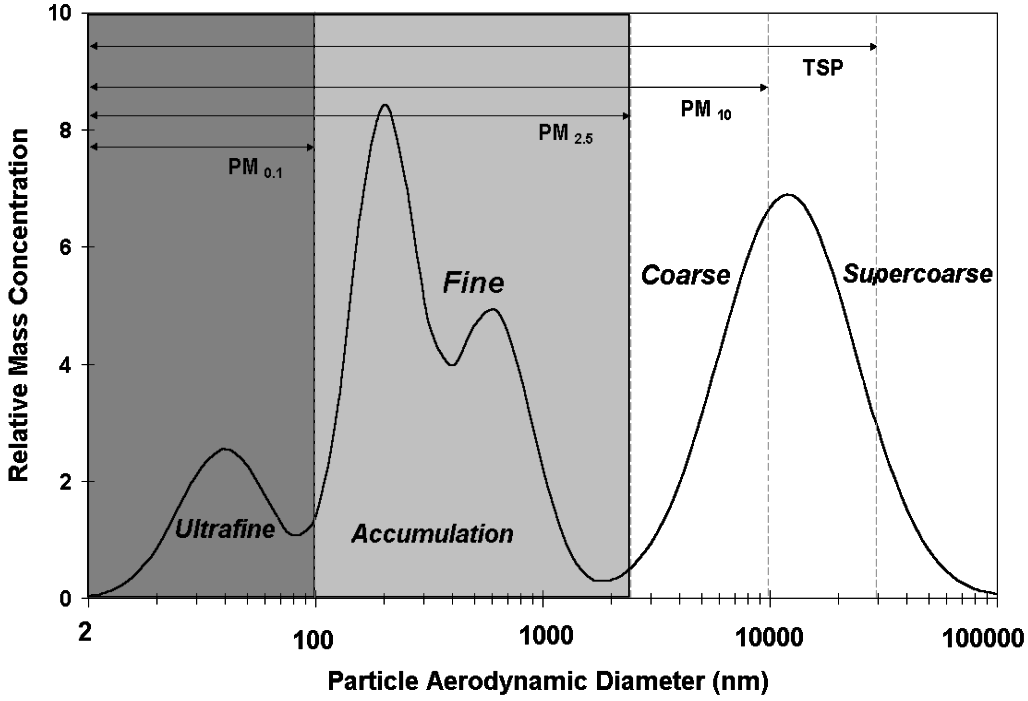


Figure 2.3.1 - Particle size distribution of atmospheric particles (adapted from Slezakova et al., 2013).

Some particles serve as nuclei upon which vapours condense and some react chemically with atmospheric gases or vapours to form different compounds. Gases, vapours and particles exist in any environment as individual molecules in random motion. When two particles collide, they tend to adhere to each other by the action of attractive surface forces, forming progressively larger particles by agglomeration. As their size enlarges as well as their mass the probability of falling to the ground increases rather than remaining airborne. This process is called sedimentation. Impactation on solid surfaces (vegetation, soil and buildings) can also help the occurrence of this phenomenon (Boubel et al., 1994).

Before the influence of human activities, particles existed in the atmosphere from natural sources. This included all the particulate forms of condensed water vapour, the condensed and reacted forms of natural organic vapours, salt particles originated by the evaporation of water from sea spray, wind-born pollen, fungi, molds, algae, yeasts, rusts, bacteria and debris from live and decaying animal and plant life, particles from volcanic and other geothermal eruption, particles wind eroded from beaches, deserts, soil, rock and from forest fires started by lightning, as well as particles entering the troposphere from outer space (Boubel et al., 1994).

Sources of particles can be either stationary (power plants, factories, etc.) or mobile (vehicles). The chemical composition, transport and destination of airborne particles are directly associated with the characteristics of the surrounding gas. Particles transported long distances can serve as a mean to transportation of adsorbed contaminants to reach water bodies, soils, plants and animals (Boubel et al., 1994).

Personal exposure to $PM_{2.5}$ can result in a decrease of heart rate variability indices, weakening autonomic function (Huang et al., 2014).

Children are of special concern in exposure to air pollutants because of their relatively smaller lung surface, their stage of health development, and the proportion of time they spend indors (Barakat-Haddad et al., 2012). Exposure to $PM_{2.5}$ has been linked to the risk of acute lower respiratory infection among young children. The increased the exposure, the higher risk of these types of infection children will develop in younger age. The reduction of indoor air pollution for neonates and young infants could be determining in preventing these scenarios (Gurley et al., 2014; Gao et al., 2009). An example of how much the exposure to particles can be grave in children, both indoor and outdoor, is a study by Bernstein et al. (2008). Authors concluded that children living near freeways experienced far more respiratory symptoms than those living further away (Bernstein et al., 2008). Also, there is a study with results that suggest that exposure to particulate matter in childhood could be indirectly related with respiratory conditions later in adulthood (Barakat-Haddad et al., 2012). Decreased lung function later in life and asthma onset could be influenced by early exposure to air pollution, as well as potentiate the influence of other exposure such as virus infections (Raaschou-Nielsen et al., 2010). Allergies and bronchial asthma in children and adults was also proven to be associated with house dust when phthalate was adsorbed in PM (Ait Bamai et al., 2014). In Japan a study concluded that there is a relationship between the increase of $PM_{2.5}$ and the number of outpatients with allergic conjunctivitis suggesting that these particles contribute to the exacerbation of this allergy (Mimura et al., 2014). Personal exposure to $PM_{2.5}$ can result in a decrease of heart rate variability indices, weakening autonomic function (Huang et al., 2014). Exposure to PM can also lead to reduced school productivity and attendance as well as input discomfort in children (Madureira et al., 2012).

2.3.1 Coarse Mode

Coarse mode particle refers to the particles with an aerodynamic diameter bigger than 2.5 μm. Mechanical processes such as wind erosion produce these particles, grinding activities as agricultural processes mining operations, wind transport of pollen and spores, particles from plant fibres and leaves and erosion of uncovered soils and unpaved roads (Tranfield and Walker, 2012). Tyre wear on the road has been considered also to contribute mainly to the formation of larger size particles (Morawska et al., 2008). Due to their large size, they only remain in the atmosphere for a few hours, depositing later because of their high mass and inertia. Particles with a diameter between 3 and 10 μm are filtrated by the upper respiratory system and cannot penetrate deep into lungs (Slezakova et al., 2013b).

As can be observed from Figure 2.3.1, the subgroup corresponding to upper fraction of coarse mode bigger than 10 μm is named as supercoarse. From health perspective, particle of this size mode cannot enter respiratory system and are considered not harmful. However, these particles have an adverse effect on the environment so the monitoring is necessary to a particle size of 30 μm, also designated as total suspended particles (TSP). Particles with diameters larger than 10 μm rarely travel long distances, and sediment shortly after their release into the atmosphere near their source.

Indoors, activities like dusting, sweeping, hovering and sitting on furniture can lead to an increase of coarse mode particles in the air. Human movements such as walking, dancing and children playing are also determinative contributors for the suspension or ressuspension (Qian et al., 2014) of these larger particles (especially if the floor is lined with any sort of carpet). Folding clothes and blankets and making a bed are also important activities as well as the presence of pets and pouring of kitty litter (Morawska et al., 2013). The type of furniture material and the high number of people in relation to room volume has also been demonstrated as relevant in raising levels of coarse particles concentration (Diapouli et al., 2007).

Frying foods in fat and liquids is also known to increase concentrations of both smaller and larger particles (Morawska et al., 2013).

These particles are typically quantified as mass per volume of air, $\mu g\ m^{-3}$ are commonly used units.

2.3.2 Fine Mode

Fine mode is subdivided in two groups: nuclei and accumulation mode. These particles are sized smaller than 2.5 μm. Nuclei mode is also designated as "ultrafine particles" with an aerodynamic diameter smaller than 0.1 μm (Slezakova et al., 2013b). These particles are emitted from combustion sources or formed by nucleation (condensation of low-vapour-pressure substances formed by high-temperature vaporization) or by chemical reactions in atmosphere with the result of the formation of new particles (nuclei). Accumulation mode refers to particles between 0.1 and 1 μm. Particles of this mode are formed either from nuclei mode ones by coagulation (most efficient for large numbers of particles) or by condensation of gas or vapour molecules on the surface of existing particles (more efficient for large surface area).

Anthropogenic sources are the main source of this range of particles. Fine particles are normally produced in industrial combustion processes and in vehicle exhaust outdoor. Indoor sources of UFP are discussed further in the next section.

Gravitational settling of fine particles take days to occur due to the small size and mass of this fraction, allowing them to be transported for long distances by wind currents.

Fine mode particles are able to enter deep into respiratory system. The inhalation of these particles can result in its deposition in the conducting airways of the lungs. Nevertheless, some are able to penetrate beyond this level entering alveolar region (Slezakova et al., 2013a).

Airborne concentrations fine are typically quantified in as particle mass per volume of air (μg m^{-3}) for $PM_{2.5}$ and in terms of number of particles per unit volume of air for UFP (commonly expressed as particle number cm^{-3}).

2.4 Nuclei Mode - Ultrafine Particles

2.4.1 Properties

Ultrafine particles are also designated as nanoparticles or belonging to nuclei mode. Some authors consider that nanoparticles relate to particles with an aerodynamic diameter smaller than 0.05 μm (Kittelson, 1998). Although the scientific community has not reached a consensus, ultrafine particles and nanoparticle are often used as equivalent in a somewhat arbitrary designation of a large size range of particles (Morawska et al., 2008).

Recent studies on ultrafine particles (with $D_{ae} \leq 0.1$ μm) have shown that particles of this size l are able to enter blood stream, penetrating cell membranes and depositing on secondary and vital organs including penetrating through the placenta to the fetus (Burtscher and Schuepp, 2012).

UFP are present in the environment resulting from fresh emissions that yet have to undergo chemical reactions and modification processes. They are found in high number concentration near their sources. They are considered highly chemically reactive, resulting from their small size but large surface area allowing the possibility of adsorbing organic pollutants resulting in the transport of great amounts of toxic air species (Bernstein et al., 2008).

2.4.2 Sources

The anthropogenic sources of UFP outdoors can be due to commercial productions and industrial emissions such as power plants, incinerators and other various processes (heating, smelting, welding, etc.). Many studies have concluded that engine combustions are the major contributor in the formation of UFP in urban environments (Morawska et al., 2008) which include vehicles, trains, ships and airplanes. Diesel-fuel engines are found to produce the highest levels of ultrafine particles. It has been shown that in urban environments the smaller particles contribute the most for the total particle number concentrations (Kittelson, 1998, Morawska et al., 2008).

Associated with engine combustion, UFP particles can be primary or secondary. Primary particle are those formed in these engines or tailpipes and emitted directly as they are in the atmosphere. Sizes of primary particles typically range from 0.03 μm to 0.5 μm and consist of a solid carbonaceous material with metallic ash (derived from lubricating oil additives and engine wear) and adsorbed or condensed hydrocarbons and sulphur compounds.

Secondary particles result from cooling and condensation of hot exhaust gases from the vehicle tailpipe, forming nuclei mode particles. The ideal conditions for the formation of secondary particles are strongly affected by the dilution conditions, temperature and residence time. Studies suggest that dilution processes that involved rapid cooling and mixing of the exhaust resulted in more nucleation mode formation concluding that particle concentration decreased when he exhaust was more diluted (Morawska et al., 2008, Kittelson, 1998).

There are numerous indoor sources that produce UFP. Cooking is the major source of indoor air pollution, especially frying and boiling but also toasting, baking, grilling and usage of electrical cooker (Abdullahi et al., 2013). However, other cooking activities can be a source of UFP. A recent study revealed that the cooking of pre-packed popcorn in microwave releases large amounts of particles, mostly in the ultrafine size range (Zhang et al., 2014). The operating mode and fan conditions are decisive parameters in particle formation mechanisms (Rim et al., 2012). Cleaning and floor polishing, burning incense and candles, smoking, lightening fire places, kerosene heating and mosquito coils as well as the operation of hard copy devices are some of the activities that contribute also to the rise of particle levels indoors. It is clear that the characterization of this sources and their usage greatly differ from house to house and is dependent on the occupants activities and patterns, resulting on particle concentration levels very disparate from home to home (Dura and Szalay, 2007).

Tobacco is a significant source of UFP, and can perpetuate high levels when smoked indoors. Tobacco smoke is known to be a suspension of liquid particles of median diameter of 0.5 μm, with a range from 0.01 to 1.0 μm, forming a superheated vapour from the burning tobacco condenses (Afshari et al., 2005). UFP resulting from tobacco burning seems to undergo particle coagulation and/or condensation or other particle growth mechanisms, since studies often detect an increase of concentration in bigger diameter size ranges after the occurrence of this source (Afshari et al., 2005).

2.4.3 Processes

UFP formation can occur in the atmosphere mainly by the following three processes (Sioutas et al., 2005):

i) Processes involving combustion which is associated specially with industrial and traffic and are emitted directly into atmosphere;
ii) Nucleation and condensation resulting from the cooling of hot supersaturated vapours emitted from combustion processes;
iii) Condensation and nucleation of low vapour pressure chemical species as a product of chemical reactions in the atmosphere.

Although the study and knowledge about the formation of UFP particles is still developing and shows significant gaps, it is possible to conclude some findings of the existing information. The concentration of non-volatile vapours is considered to be the driving force for nucleation and

particle growth (Kulmala et al., 2004). Therefore, there is a strong relationship between UFP formation and sulphuric acid and other low volatility gases, particularly organic compounds and iodine vapours. Solar radiation and atmospheric mixing processes seem to influence greatly the formation of aerosols in the atmosphere (Kulmala et al., 2004).

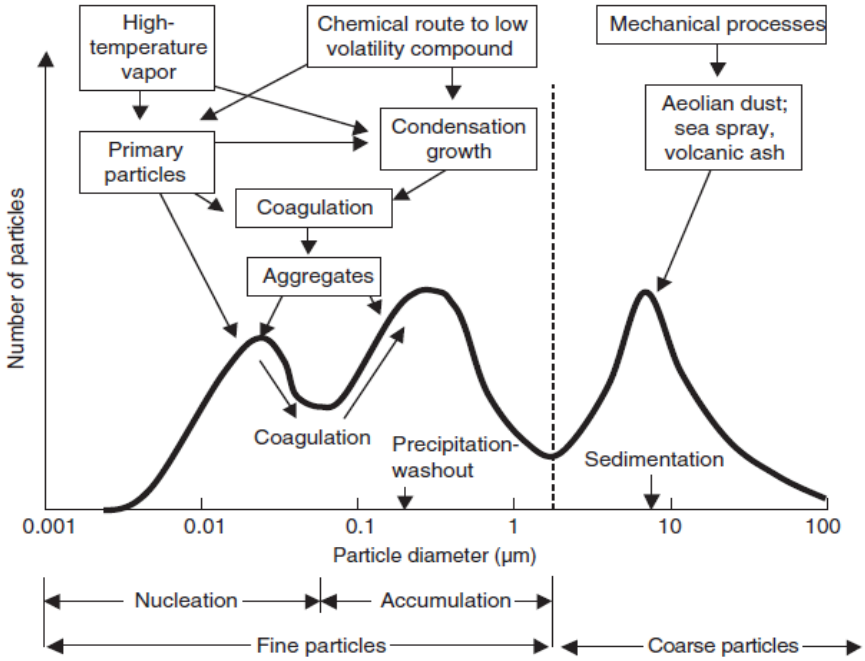


Figure 2.4.1- Schematic size distribution of tropospheric particles crossed with selected sources and pathways of particle formation (adapted from Boubel et al., 1994).

Particles formations in continental boundary level have been also observed, often called "regional nucleation events". These events have been found in a variety of environments like forest areas (Kavouras et al., 1999), rural and remote continental sites, urban centres including highly polluted areas and also in marine boundary layer, although the last is very rare. Photochemistry plays an important role in aerosols formation since it is only observed during daytime. There is also evidence that particle formation rates are higher in the summer, supporting the hypotheses that photochemistry has a strong influence on producing vapours that lead to nucleation of particles (Weber et al., 1997). Other parameters that have shown signs of influence in the formation of particles are low relative humidity, high vapour source rate and low pre-existing particle concentration (Kulmala et al., 2004). Very small ultrafine particles cannot be observed due to instrumental limitations (Kulmala et al., 2004). Particle formation occurs with nucleation. After formation, particle growth occurs through coagulation and condensation. Coagulation happens when particles collide and adhere to each other, while condensation is a function of saturated vapour pressure and particle surface area available, also

dependent on particle size (Holmes, 2007, Boubel et al., 1994). The vast majority of studies reported particle growth rates in the range of 1-10 nm h^{-1} implicating that it takes from 0.5 to 3 days before nucleated particles behave as cloud condensation nuclei (Kulmala et al., 2004). In terms of nucleation processes in the environment, there is not much certainty on it development. The most probable nucleation processes are (Kulmala and Kerminen, 2008):

i) Homogeneous binary water-sulphuric acid nucleation;
ii) Homogeneous ternary water-sulphuric acid-ammonia nucleation;
iii) Ion-induced nucleation of binary (water sulphuric acid) or ternary inorganic vapours or of organic vapours;
iv) Barrier-less homogeneous nucleation.

Homogeneous binary water-sulphuric acid nucleation is related with industrial plumes and free troposphere and possibility. Ternary water-sulphuric acid-ammonia nucleation is linked to continental boundary layer. Ion-induced nucleation of binary or ternary inorganic vapours or of organic vapours occur in upper troposphere and lower stratosphere whereas barrier-less homogeneous nucleation refers to coastal environments. However, future research on this topic and field measurements is necessary in order to better understand the phenomenon of particle formation that takes place in a variety of different scenarios all over the world (Kulmala and Kerminen, 2008).

2.4.4 Composition

The composition of UFP is very variable and dependent on its source and formations. Information on particle composition allows understanding about its source. Receptor models use chemical composition and morphology of particles as means to identify source of the particles (Boubel et al., 1994).

There is very little information available on the composition of UFP particles. It is recognized that its chemical composition is related with both particle size and shape. In fact, the presence of toxic compounds adsorbed in particle surface, such as heavy metals and polycyclic aromatic hydrocarbons can cause many preoccupying adverse effects to human health.

Composition of UFP may include inorganic compounds such as sulphates, chloride, nitrates, sulphates and trace metals (Slezakova et al., 2013b).

Metals in particles have their origin in natural or anthropogenic sources. There can have many types of metal in particles depending on its source such as zinc, iron, lead, arsenic, manganese, etc. Natural origin can be related with oceanic aerosols, volcanic dust or soil crust while anthropological sources can be related with power plants, emissions from motor engines, waste incinerators and high temperature industrial processes (Slezakova, 2009).

Elemental and organic carbon, volatile and semi-volatile organic compounds, crystal materials and biological components may also be a part of UFP composition. Studies suggest that in some environments organic material such as polycyclic aromatic hydrocarbons were the most abundant portion of atmospheric ultrafine particle (Slezakova et al., 2013b). However, the urge to conduct more studies in the composition of these particles is evident; in order to better understand chemistry dynamics of ultrafine particles.

2.4.5 Environmental Effects

Aerosols have an effect on radiation intensity, since it creates a barrier preventing radiation distribution on earth surface as well as indirectly altering the scattering characteristics of clouds influencing negatively terrestrial carbon sink, thus contributing to the rise of global climate (Kulmala and Kerminen, 2008, Weber et al., 1997).

Particles contribute to the formation of acid rain, since they can serve as vehicle in the transportation of chemicals contaminants. They can enter ecosystems and potentially reduce the pH of receiving water, as well lowering the pH of the rain in the process of being washed out from the atmosphere (Boubel et al., 1994). Compounds with an elevated K_{OC} that are highly sorptive can be transported in particles for a long-range distance (Boubel et al., 1994).

2.4.6 Health Effects

Although outdoor air pollution remains as a concern, there is a growing recognition that indoor air pollution is of equal or greater significance to human health. The amount of time people spend indoors, the wide and varied range of indoor emission sources and the higher levels of some pollutants concentration indoor compared to outdoors are some of the reasons for this occurrence. The existence of clean indoor air is of great importance particularly for susceptible groups like infants, children and the elderly, including also people with genetic predisposal to disease and people that have developed respiratory or cardiovascular diseases. Health effects

of particles are strongly linked to its size since this characteristic is determinant of the region in the organism where these particles will deposit (Dura and Szalay, 2007; Morawska et al., 2008).

UFP have a behavior similar to a gas, entering all parts of the lung. These nanoparticles have the ability to deposit in the alveolar surface and be transported through the bloodstream or lymphatic system to vital organs (Burtscher and Schuepp, 2012). The place of particle deposition depends on the particle size, particle mass concentration, molecular composition, pH, and solubility, the fact that the individual is a smoker or non-smoker or has a lung-associated disease (Sunyer et al., 2000). Compared to cellular structure size, UFP are very small. This fact is very important in the problems they may represent to the lung. (Tan et al., 2014; Slezakova et al., 2013a; Donaldson et al., 2001)

Combustion processes, volatile organic compounds and biological pollutants are also responsible for the aggravation of respiratory and cardiovascular diseases (Dura and Szalay, 2007). Developing countries have a higher exposure to indoor air pollution, since modernization of energy sources for cooking purposes is still much delayed in relation to developed countries, with more than half of the world's population relying on biomass combustion to meet their domestic energy needs (Bruce et al., 2000; Clark et al., 2010). Studies from developing countries on health effects of household pollution demonstrate how much indoor air quality determines occupants' wellbeing. Women, relating to a preference of gender in cooking activities in many cultures, show a higher dose of exposure resulting in more negative consequences on their health, relatively to men (Buonanno et al., 2014). Chronic obstructive pulmonary disease (COPD) in women and men has been associated with indoor air pollution caused by domestic use of solid fuel like coal and biomass (Gao et al., 2009). It was demonstrated than indoor air pollution was responsible for the development of lung cancer among several non-smoker females (Mu et al., 2013).

It is important to take into account that the exposure to air pollution represents an exposure to a mixture of pollutants. This combination of pollutants can lead to synergistic effects. Furthermore, the combination of some pollutants allied to other factors like ventilation rate can lead to effects not so well known and studied. For example, the exposure to indoor burning of biomass allied to poorly ventilated buildings (a typical scenario in developing countries) can lead to cataract formation.

Therefore, in the indoor environment, it is important to focus not only in individual pollutants but on reducing their sources (WHO, 2010).

2.5 Legislation

For the European Union (EU), indoor climate environment advising are in Directive 2010/31/EU, which promotes the improvement of energy performance on buildings within EU, taking into account outdoor climatic and local conditions, as well as indoor climate requirements and cost-effectiveness. It includes the common general framework for methodology on calculation the integrated energy performance of buildings and building units, the application of minimum requirements to the energy performance of new and other buildings (EU Directive, 2010).

This Directive was approved and transposed to Portuguese legislation in Decreto-Lei 118/2013. It promotes and ensures the improvement of energetic performance of buildings, including other relevant regulations on the topic (Decreto-Lei nº 118/2013).

Portaria 353-A.2013 establishes the minimum values for new airflow by space as well as protection thresholds and reference conditions for indoor air pollutants for new, subjected to big interventions and existing commercial and services buildings, and respective evaluation methodology. These can be consulted in the Table 2.5.1.

Table 2.5.1 - Portuguese threshold limits for indoor pollutants and tolerance margin.

Pollutants	***Protection threshold***	***Units***	***Tolerance margin (%)***
Suspended particles (PM_{10})	50	$\mu g\ m^{-3}$	100
Suspended particles ($PM_{2.5}$)	25	$\mu g\ m^{-3}$	100
Total volatile organic compounds (VOC's)	600	$\mu g\ m^{-3}$	100
Carbon monoxide (CO)	10	$mg\ m^{-3}$	-

	9	ppmv	
Formaldehyde (Ch_2O)	100	$\mu g\ m^{-3}$	-
	0,08	ppmv	
Carbon dioxide (CO_2)	2250	$mg\ m^{-3}$	30
	1250	ppmv	
Radon	400	$Bq\ m^{-3}$	-

2.6 References

ABDULLAHI, K. L., DELGADO-SABORIT, J. M. & HARRISON, R. M. 2013. Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: A review. *Atmospheric Environment,* 71**,** 260-294.

AIT BAMAI, Y., SHIBATA, E., SAITO, I., ARAKI, A., KANAZAWA, A., MORIMOTO, K., NAKAYAMA, K., TANAKA, M., TAKIGAWA, T., YOSHIMURA, T., CHIKARA, H., SAIJO, Y. & KISHI, R. 2014. Exposure to house dust phthalates in relation to asthma and allergies in both children and adults. *Science of The Total Environment,* 485–486**,** 153-163.

APA. 2014. *Agência Portuguesa do Ambiente - O que é a qualidade do ar interior?* [Online]. Available: http://www.apambiente.pt/index.php?ref=16&subref=82&sub2ref=319&sub3ref=338 [Accessed 28/05/2014.

ALVES, S. & MARIANO, F. 2014. Paris limita circulação devido a perigoso nível de poluição. *Jornal de Notícias - Online version* - http://www.jn.pt/PaginaInicial/Mundo/Interior.aspx?content_id=3754197&page=-1 - 15/03/2014BARAKAT-HADDAD, C., ELLIOTT, S. J. & PENGELLY, D. 2012. Health Impacts of Air Pollution: A Life Course Approach for Examining Predictors of Respiratory Health in Adulthood. *Annals of Epidemiology,* 22**,** 239-249.

BERNSTEIN, J. A., ALEXIS, N., BACCHUS, H., BERNSTEIN, I. L., FRITZ, P., HORNER, E., LI, N., MASON, S., NEL, A., OULLETTE, J., REIJULA, K., REPONEN, T., SELTZER, J., SMITH, A. & TARLO, S. M. 2008. The health effects of nonindustrial indoor air pollution. *Journal of Allergy and Clinical Immunology,* 121**,** 585-591.

BOUBEL, R. W., FOX, D. L., TURNER, D. B. & STERN, A. C. 1994. Fundamentals of air pollution.

BRUCE, N., PEREZ-PADILLA, R. & ALBALAK, R. 2000. Indoor air pollution in developing countries: a major environmental and public health challenge. *Bulletin of the World Health Organization,* 78**,** 1078-1092.

BUONANNO, G., STABILE, L. & MORAWSKA, L. 2014. Personal exposure to ultrafine particles: The influence of time-activity patterns. *Science of the Total Environment,* 468**,** 903-907.

BURTSCHER, H. & SCHUEPP, K. 2012. The occurrence of ultrafine particles in the specific environment of children. *Paediatric Respiratory Reviews,* 13**,** 89-94.

CLARK, M. L., REYNOLDS, S. J., BURCH, J. B., CONWAY, S., BACHAND, A. M. & PEEL, J. L. 2010. Indoor air pollution, cookstove quality, and housing characteristics in two Honduran communities. *Environmental Research,* 110**,** 12-18.

DECRETO-LEI Nº 118/2013 Certificação Energética e Qualidade do Ar Interior.

DIAPOULI, E., CHALOULAKOU, A. & SPYRELLIS, N. 2007. Levels of ultrafine particles in different microenvironments—implications to children exposure. *Science of the Total Environment,* 388**,** 128-136.

DIMITROULOPOULOU, C. 2012. Ventilation in European dwellings: A review. *Building and Environment,* 47**,** 109-125.

DONALDSON, K., STONE, V., CLOUTER, A., RENWICK, L. & MACNEE, W. 2001. Ultrafine particles. *Occupational and environmental medicine,* 58**,** 211-216.

DONGHYUN, R. & NOVOSELAC, A. 2010. Occupational Exposure to Hazardous Airborne Pollutants: Effects of Air Mixing and Source Location. 7**,** 683-92.

DURA, G. & SZALAY, B. 2007. Particle exposure through the indoor air environment. *In:* SIMEONOVA, P. P., OPOPOL, N. & LUSTER, M. I. (eds.) *Nanotechnology - Toxicological Issues and Environmental Safety.* Dordrecht: Springer.

EPA. 2012. *United States Envionment Protection Agency - IAQ in Homes* [Online]. Available: http://www.epa.gov/iaq/homes/index.html [Accessed 24/04/2014.

EU DIRECTIVE 2010. DIRECTIVE 2010/31/EU OF THE EUROPEAN PARLIMENT AND OF THE COUNCIL of 19 May 2010 on energy performance of buildings (recast). *In:* THE EUROPEAN PARLIMENT AND THE COUNCIL OF THE EUROPEAN UNION (ed.).

FRANKLIN, P. J. 2007. Indoor air quality and respiratory health of children. *Paediatric Respiratory Reviews,* 8**,** 281-286.

GALL, E. T., CARTER, E. M., EARNEST, C. M. & STEPHENS, B. 2013. Indoor Air Pollution in Developing Countries: Research and Implementation Needs for Improvements in Global Public Health. *American Journal of Public Health,* 103**,** e67-e72.

GAO, X., YU, Q., GU, Q., CHEN, Y., DING, K., ZHU, J. & CHEN, L. 2009. Indoor air pollution from solid biomass fuels combustion in rural agricultural area of Tibet, China. *Indoor Air,* 19**,** 198-205.

GURLEY, E. S., SALJE, H., HOMAIRA, N., RAM, P. K., HAQUE, R., PETRI, W. A., BRESEE, J., MOSS, W. J., LUBY, S. P., BREYSSE, P. & AZZIZ-BAUMGARTNER, E. 2014. Indoor Exposure to Particulate Matter and Age at First Acute Lower Respiratory Infection in a Low-Income Urban Community in Bangladesh. *American Journal of Epidemiology,* 179**,** 967-973.

HOLMES, N. S. 2007. A review of particle formation events and growth in the atmosphere in the various environments and discussion of mechanistic implications. *Atmospheric Environment,* 41**,** 2183-2201.

HUANG, Y.-L., CHEN, H.-W., HAN, B.-C., LIU, C.-W., CHUANG, H.-C., LIN, L.-Y. & CHUANG, K.-J. 2014. Personal Exposure to Household Particulate Matter, Household Activities and Heart Rate Variability among Housewives. *PLoS ONE,* 9**,** 1-5.

HULIN, M., CAILLAUD, D. & ANNESI-MAESANO, I. 2010. Indoor air pollution and childhood asthma: variations between urban and rural areas. *Indoor Air,* 20**,** 502-514.

KAVOURAS, I. G., MIHALOPOULOS, N. & STEPHANOU, E. 1999. Formation and gas/particle partitioning of monoterpenes photo-oxidation products over forests. *Geophysical Research Letters,* 26**,** 55-58.

KITTELSON, D. B. 1998. Engines and nanoparticles: a review. *Journal of Aerosol Science,* 29**,** 575-588.

KULMALA, M. & KERMINEN, V.-M. 2008. On the formation and growth of atmospheric nanoparticles. *Atmospheric Research,* 90**,** 132-150.

KULMALA, M., VEHKAMÄKI, H., PETÄJÄ, T., DAL MASO, M., LAURI, A., KERMINEN, V. M., BIRMILI, W. & MCMURRY, P. H. 2004. Formation and growth rates of ultrafine atmospheric particles: a review of observations. *Journal of Aerosol Science,* 35**,** 143-176.

MADUREIRA, J., PACIÊNCIA, I. & FERNANDES, E. D. O. 2012. Levels and indoor–outdoor relationships of size-specific particulate matter in naturally ventilated Portuguese schools. *Journal of Toxicology and Environmental Health, Part A,* 75**,** 1423-1436.

MIMURA, T., ICHINOSE, T., YAMAGAMI, S., FUJISHIMA, H., KAMEI, Y., GOTO, M., TAKADA, S. & MATSUBARA, M. 2014. Airborne particulate matter (PM2.5) and the prevalence of allergic conjunctivitis in Japan. *Science of The Total Environment,* 487**,** 493-499.

MORAWSKA, L., AFSHARI, A., BAE, G. N., BUONANNO, G., CHAO, C. Y. H., HANNINEN, O., HOFMANN, W., ISAXON, C., JAYARATNE, E. R., PASANEN, P., SALTHAMMER, T., WARING, M. & WIERZBICKA, A. 2013. Indoor aerosols: from personal exposure to risk assessment. *Indoor Air,* 23**,** 462-487.

MORAWSKA, L., RISTOVSKI, Z., JAYARATNE, E., KEOGH, D. U. & LING, X. 2008. Ambient nano and ultrafine particles from motor vehicle emissions: characteristics, ambient processing and implications on human exposure. *Atmospheric Environment,* 42**,** 8113-8138.

MU, L. N., LIU, L., NIU, R. G., ZHAO, B. X., SHI, J. P., LI, Y. L., SWANSON, M., SCHEIDER, W., SU, J., CHANG, S. C., YU, S. Z. & ZHANG, Z. F. 2013. Indoor air pollution

and risk of lung cancer among Chinese female non-smokers. *Cancer Causes & Control,* 24**,** 439-450.

PALLISER, J. 2011. A breath of fresh air: Addressing indoor air quality. *Science Scope,* 35**,** 14-19.

QIAN, J., PECCIA, J. & FERRO, A. R. 2014. Walking-induced particle resuspension in indoor environments. *Atmospheric Environment,* 89**,** 464-481.

RAASCHOU-NIELSEN, O., HERMANSEN, M. N., LOLAND, L., BUCHVALD, F., PIPPER, C. B., SORENSEN, M., LOFT, S. & BISGAARD, H. 2010. Long-term exposure to indoor air pollution and wheezing symptoms in infants. *Indoor Air,* 20**,** 159-167.

RAMANATHAN, V. & FENG, Y. 2009. Air pollution, greenhouse gases and climate change: Global and regional perspectives. *Atmospheric Environment,* 43**,** 37-50.

RIBEIRO, D. 2014. Alerta máximo à poluição em Paris *Expresso - Online version*. http://expresso.sapo.pt/alerta-maximo-a-poluicao-em-paris=f860788 - 14/03/2014

RIM, D., GREEN, M., WALLACE, L., PERSILY, A. & CHOI, J.-I. 2012. Evolution of ultrafine particle size distributions following indoor episodic releases: Relative importance of coagulation, deposition and ventilation. *Aerosol Science and Technology,* 46**,** 494-503.

RIM, D., WALLACE, L. A. & PERSILY, A. K. 2013. Indoor Ultrafine Particles of Outdoor Origin: Importance of Window Opening Area and Fan Operation Condition. *Environmental Science & Technology,* 47**,** 1922-1929.

SIOUTAS, C., DELFINO, R. J. & SINGH, M. 2005. Exposure Assessment for Atmospheric Ultrafine Particles (UFPs) and Implications in Epidemiologic Research. *Environmental Health Perspectives,* 113**,** 947-955.

SLEZAKOVA, K. 2009. *Suspended Particles in Outdoor and Indoor Air: Characterization to Support Epidemiological Studies.* Philosophy Doctor, Faculdade de Engenharia da Universidade do Porto.

SLEZAKOVA, K., MORAIS, S. & PEREIRA, M. D. C. 2013a. *Atmospheric Nanoparticles and Their Impacts on Public Health*.

SLEZAKOVA, K., MORAIS, S. & PEREIRA, M. D. C. 2013b. Air Pollution: Particulate Matter. *Encyclopedia of Environmental Management.* Taylor & Francis.

SPILAK, M. P., FREDERIKSEN, M., KOLARIK, B. & GUNNARSEN, L. 2014. Exposure to ultrafine particles in relation to indoor events and dwelling characteristics. *Building and Environment,* 74**,** 65-74.

SUNYER, J., SCHWARTZ, J., TOBÍAS, A., MACFARLANE, D., GARCIA, J. & ANTÓ, J. M. 2000. Patients with chronic obstructive pulmonary disease are at increased risk of death associated with urban particle air pollution: a case-crossover analysis. *American Journal of Epidemiology,* 151**,** 50-56.

TAN, J.-H., DUAN, J.-C., CHAI, F.-H., HE, K.-B. & HAO, J.-M. 2014. Source apportionment of size segregated fine/ultrafine particle by PMF in Beijing. *Atmospheric Research,* 139**,** 90-100.

TRANFIELD, E. M. & WALKER, D. C. 2012. Understanding Human Illness and Death Following Exposure to Particulate Matter Air Pollution. *Environmental Health–Emerging Issues and Practice***,** 81-102.

VIEGI, G., SIMONI, M., SCOGNAMIGLIO, A., BALDACCI, S., PISTELLI, F., CARROZZI, L. & ANNESI-MAESANO, I. 2004. Indoor air pollution and airway disease [State of the Art]. *The International Journal of Tuberculosis and Lung Disease,* 8**,** 1401-1415.

WEBER, R., MARTI, J., MCMURRY, P., EISELE, F., TANNER, D. & JEFFERSON, A. 1997. Measurements of new particle formation and ultrafine particle growth rates at a clean continental site. *Journal of Geophysical Research: Atmospheres (1984–2012),* 102**,** 4375-4385.

WHO 2006. Air quality guidelines global update 2005: Particulate matter, ozone, nitrogen dioxide and sulfor dioxide - Summary of risk assessment. *In:* WORLD HEALTH ORGANIZATION (ed.). Copenhagen: World Health Organization Regional Office for Europe,.

WHO 2010. WHO guidelines for indoor air quality: selected pollutants. *In:* WHO REGIONAL OFFICE FOR EUROPE (ed.). Copenhagen: WHO European Centre for Environment and Health,.

WHO 2014. Burden of disease from Household Air Pollution 2012 - Summary of results. Geneva: World Health Organization.

ZHANG, L.-L., WEI, F.-S. & WU, G.-P. Study on the relationship between lung cancer of indoor air pollution exposure and trace elements in blood plasma. 2009 Piscataway, NJ, USA. IEEE, 4 pp.

ZHANG, Q., AVALOS, J. & ZHU, Y. 2014. Fine and ultrafine particle emissions from microwave popcorn. *Indoor Air,* 24**,** 190-198.

3. Materials and Methods

3.1 Homes Characterization

Four different homes were evaluated in this study: three homes were located in the area of "Grande Porto" (two in Porto and one in Matosinhos) and the fourth home was located in Resende County. They are further characterized in the following sections.

3.1.1 Home 1

Home 1 (H1) was an apartment on the 4th floor of a multiunit building, located in Coronel Almeida Valente Street, 4200-033, in Paranhos municipality, Porto, Portugal.

This home was situated in Asprela university center (7 public faculties belonging to University of Porto, 2 public institutes of Polytechnic of Porto and 2 private universities). There are three major roads: VCI, Circunvalação and A3 highway. S. João Hospital and Instituto Português de Oncologia are also located in this area as well as other investigation and development centers take place in Asprela center. It is estimated that 45,000 people travel daily to this area of Porto city.

Paranhos municipality has a population of 44,290 habitants (INE, 2012).

Geographical location of H1 is presented in Figure 3.1.1. Two streets surround it: Coronel de Almeida Valente Street and Conde Aurora Street.

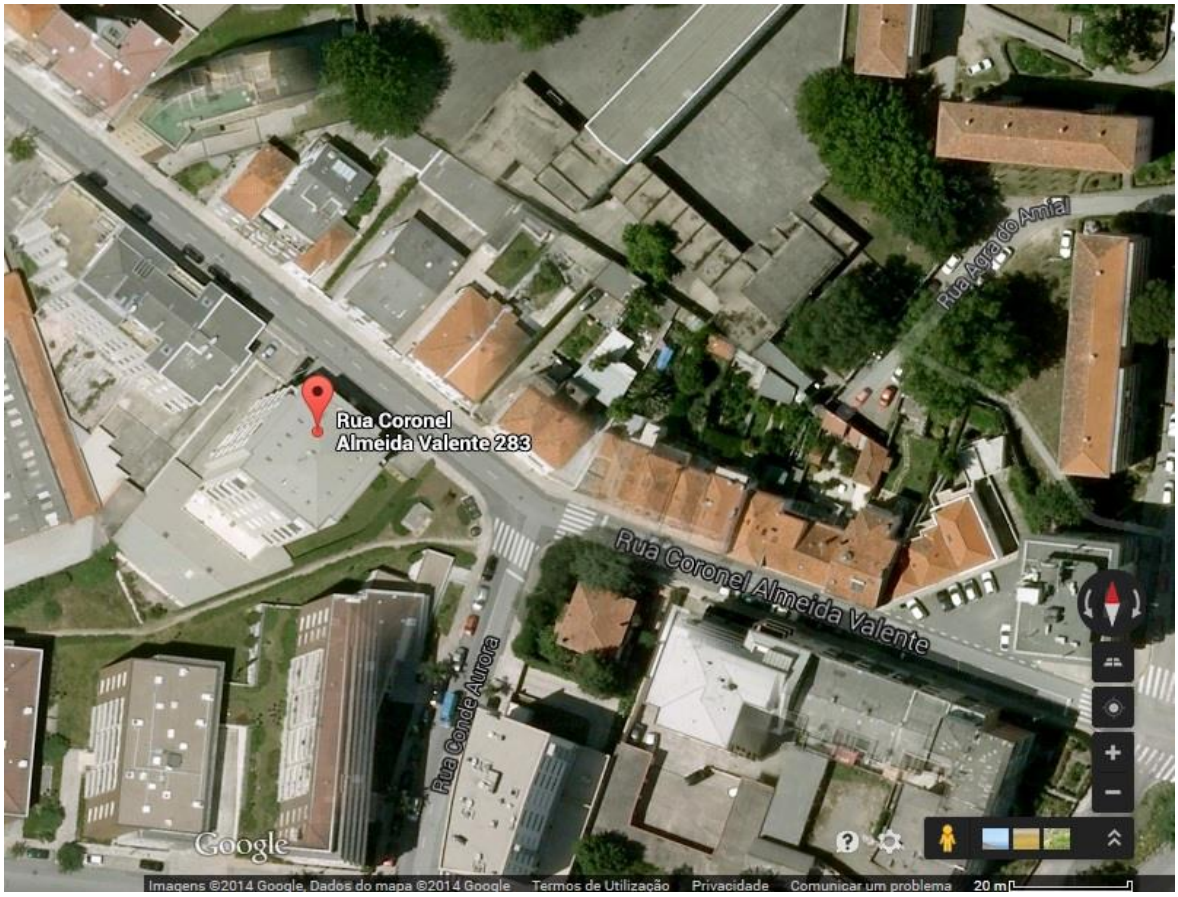


Figure 3.1.1 - Geographical layout of H1.

The construction of this building was concluded in 1974. The apartment was renovated in 1997, and in 2012 the balcony and the kitchen were remodeled (including the kitchen exhaust system).

Four people inhabit this apartment: two adults (35 and 63 years old) and two children (between 5 to 6 years old).

The sampling was carried out in the room used as in the dining and living room, which was directly connected to kitchen (Annex A.1).

In order to understand the traffic density within this area during different periods of the day, the number of cars were manually counted each hour (from 4 a.m. to 12 p.m.) for a duration of 10 minutes considering passenger cars, heavy vehicles/busses, and motorcycles. This procedure was performed during two consecutive weekdays (avoiding Mondays and Fridays). The collection of this data enabled the calculation of the average number of cars in each street surrounding the building and to establish its daily traffic density profile. This information is presented in Table 3.1.1 and Figure 3.1.2.

Table 3.1.1 - Average values for traffic density of the two streets surrounding H1.

	Coronel de Almeida Valente St		***Conde de Aurora St***	
Daily Average (vehicle min^{-1})	2.0	2.2	0.4	0.5
Street Average (vehicle min^{-1})	2.1		0.4	

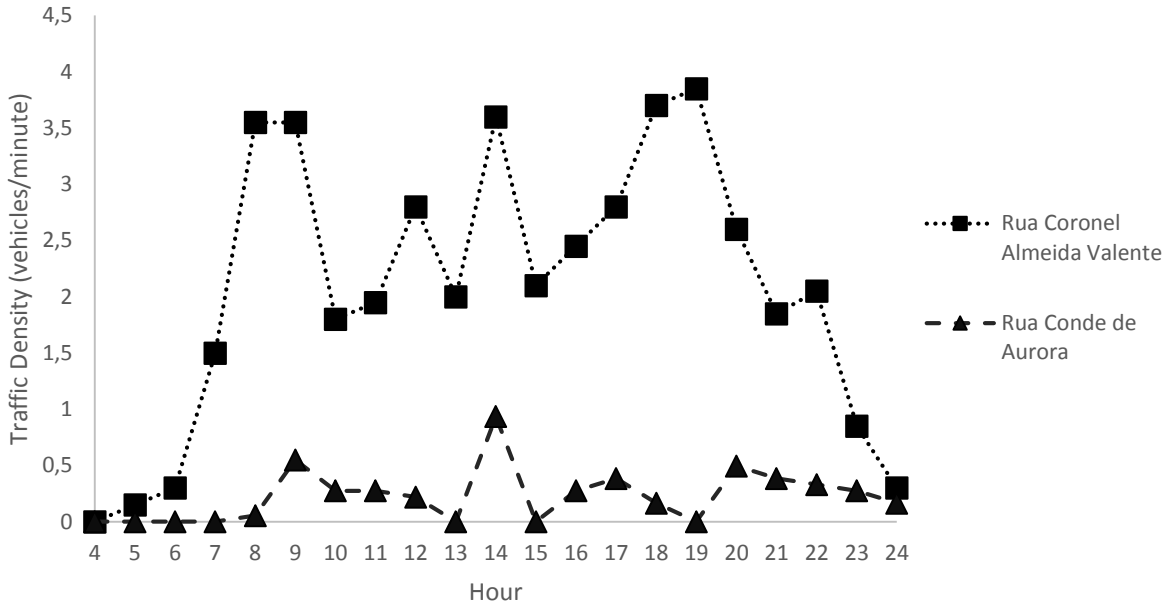


Figure 3.1.2 - Average traffic density profiles for the two streets surrounding H1.

As shown in Figure 3.3 Coronel Almeida Valente St presents a higher density than Conde Aurora St. This is related probably by the fact that Conde de Aurora St mainly gives access to local buildings and parking spaces, while Coronel Valente St connects to educational/offices buildings nearby and eventually leads to VCI.

In both streets, it can clearly see higher vehicle density in three decisive parts of the day corresponding to morning, lunch and dinner periods where private transportation takes significant importance in these periods. Overall profiles in both streets exhibited maximum and minimum values during similar daily hours.

3.1.2 Home 2

Home 2 (H2) was a house located in Nova de S. Gens Street, 4460-778 Custóias, Matosinhos, Portugal.

H2 is located in Custóias village belonging to Matosinhos County. Custóias has an area of 5.78 km^2 and has a population of 18 650 habitants (INE, 2012). It is located in 5 km to Matosinhos and 8 km to Porto. A4 highway and VRI are connected to Custóias. Five pre-secondary schools are located in Custóias' area. With a historical background connected to agriculture, Custóias economical activities have been changing to industry and services throughout time. Custóias is known for fair of local fresh products that attracts a significant number of regular customers during every weekend.

H2 is three floor house dated from 1994 (year finished) originally dimensioned for a single family with two children and has been habited to the present date. In 2012 wall painting work were conducted in in the 2nd floor. In the present, two adults and one child occupy this home: the two adults with 60 and 62 years and the child with 6 years old.

The sampling equipment was placed on the ground floor in in the living room, once again used also as dining room. This room was not directly connected to kitchen (Appendix A.2).

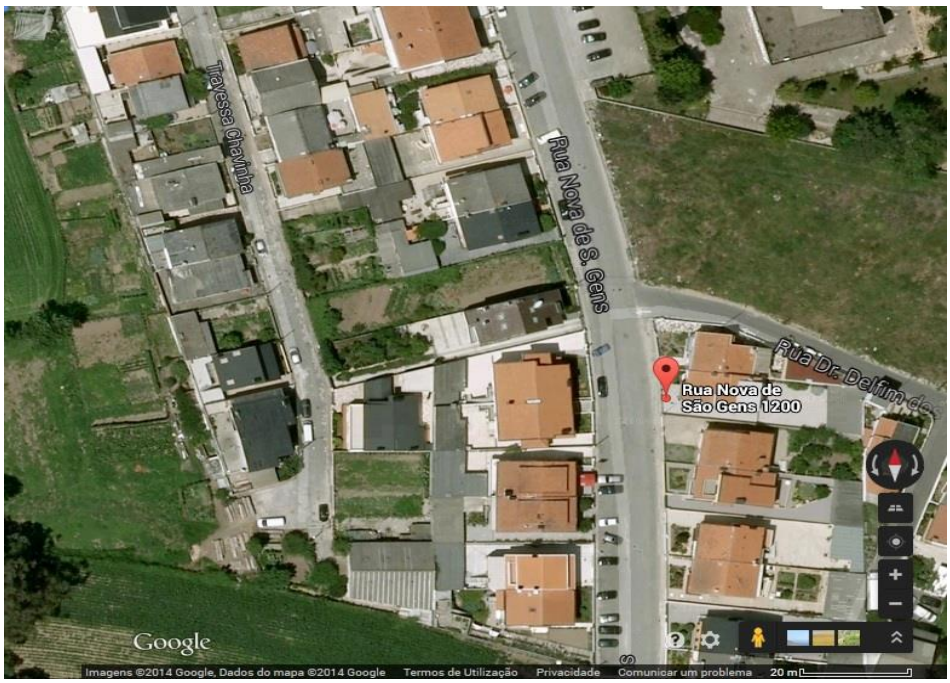


Figure 3.1.3- Geographical layout for H2.

In this case, only Nova de S. Gens Street was considered for the characterization of this home since H2 has no direct contact with Delfim dos Santos Street because of multiple dwellings are situated in between.

In order to evaluate the traffic density in this road, the same methodology as for H1 was applied to this environment from 4 a.m. to 12 p.m. in two consecutive days of the week. The average number of vehicles per minute and the average traffic density daily profiles are presented, respectively, in Table 3.1.2 and Figure 3.1.4.

Table 3.1.2 - Average values for traffic density of the street surrounding H2.

	Nova de S. Gens St	
Daily Average (vehicle min^{-1})	3.6	3.6
Street Average (vehicle min^{-1})	3.6	

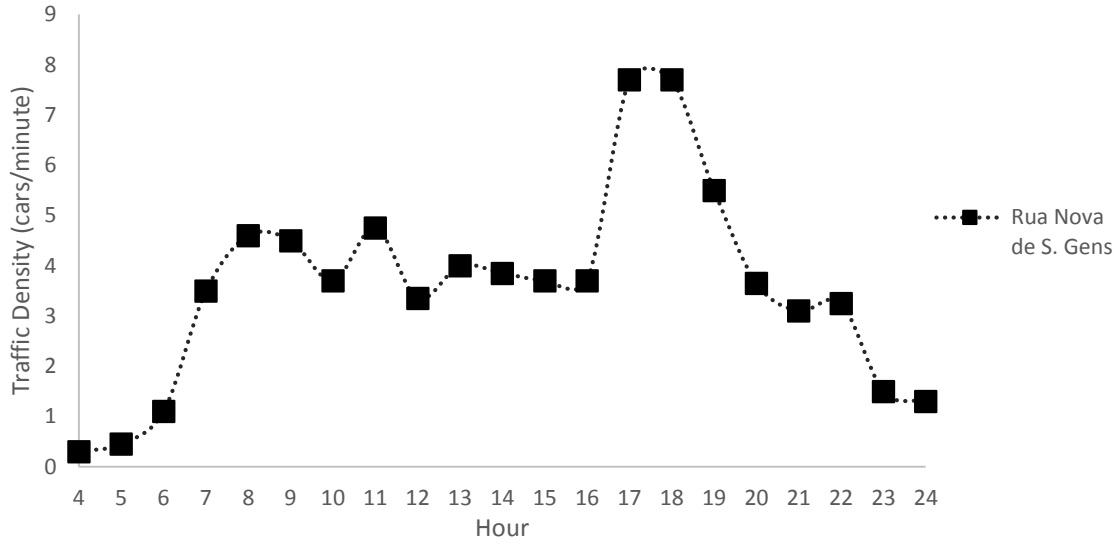


Figure 3.1.4 - Average traffic density profile for the street surrounding H2.

In Figure 3.1.4 we can see that traffic density increases from 6 a.m. until 10 a.m. representing a very broad morning peak. Around 11 a.m., it is shown another peak, probably due to lunch hour. After 1 p.m. to 4 p.m., the number of vehicles slowly decreases. The highest peak between 5 and 6 p.m. is probably because of the presence of school in this street; when classes finish at these hours parents pick up children after school. However, this occurrence also could be due to the fact that at this hour many heavy vehicles park nearby.

Since Nova de S. Gens St is one of the main roads in Custóias village and gives access eventually to A4 and VRI, (as shown in Figure 3.6) that traffic intensity is constantly relatively high during all day.

3.1.3 Home 3

Home 3 (H3) was located in Cervantes Street in a multiunit building in the 4th floor, 4050-186, Cedofeita, Porto.

Cedofeita has a population of 22,077 habitants (INE, 2012). UH3 was located near Damião de Góis Steet that gives access to the center of Porto city.

The construction of this building was finished in the year 1981 and it was not remodeled since.

The sampling in this room took place in the living room of the apartment that also is used as dining room. This division was not directly connected with the kitchen. Four people inhabit H3: two adults, one adolescent and one child.

The geographical layout of H3 is represented in the following Figure 3.1.5.

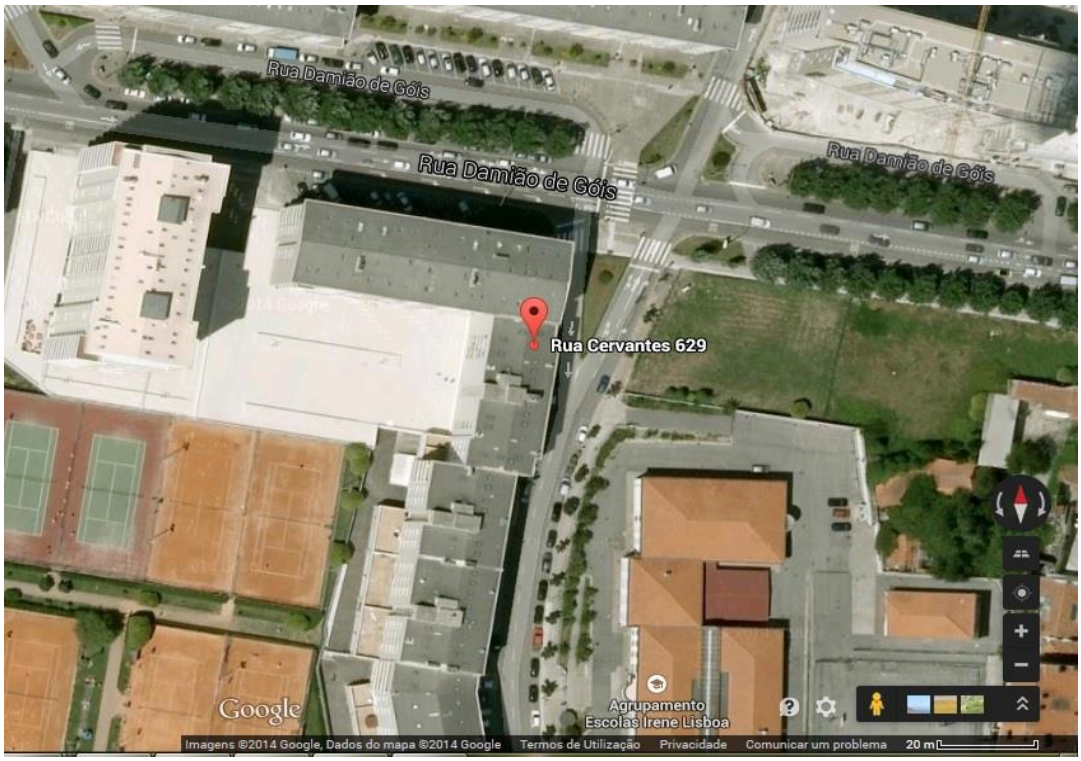


Figure 3.1.5 - H3 geographical layout.

3.1.4 Home 4

Home 4 (H4) was a house located in Tílias Avenue, number 91, 4660-013 Anreade, Resende, Portugal.

This place, known as Caldas de Aregos, refers to the population living in this specific "amphitheater" shape curve of Douro river. It refers to a part of territory of Miomães and a part of Anreade, both belonging to Resende County. H4 is placed in Anreade part of the territory. Da Cesta Brook divides the two Freguesias, passing throw. Tílias Avenue towards Douro River. S. Miguel de Anreade has a population of 1,114 habitants (INE, 2012) and the nearest city is Lamego, 38 km of distance.

Caldas de Aregos is located 90 km away from Porto. Its main economical attraction is the vastly known spa of hot springs (sulfurous bicarbonate sodium) from geothermic activity. Natural beauty of the area, rural pure and quiet atmosphere and aquatic sports by the river also attract visitors, mainly in summer.

The only main road with significant traffic is the Nacional - 222 that connects Vila Nova de Gaia to Almendra (Vila Nova de Foz Côa) and passes through Caldas the Aregos allowing the access to the closest village, Resende.

H4 was occupied by 2 adults aged 58 and 61 years and a 5 year old child. Both adults were smokers. It is necessary to remark that H4 was the only home where its inhabitants were active smokers.

The sampling took place in the living/dining of the house located in 1st floor of the building with direct connection with the kitchen (see Appendix A.4).

Figure 3.1.6 - Home 4 geographic layout.

Similarly to H1 and H2, on two consecutive days of the week on traffic density of Tílias Avenue was evaluated in order to understand the impacts of traffic emissions on the surroundings of this residence. The results are presented in Table 3.1.3; the average traffic density profile is represented in Figure 3.1.7. Since H4 is located near the Douro River, the possibility of boat traffic influence on air pollution was also considered. Although in summer period many recreational boats are known to cross this part of river, UFP sampling was performed before period of vacation and bathing season, and no boats were registered during this time.

Table 3.1.3 - Average values for traffic density of the street surrounding H4.

	Tílias Av	
Daily Average (vehicle min^{-1})	0.1	0.1
Street Average (vehicle min^{-1})	0.1	

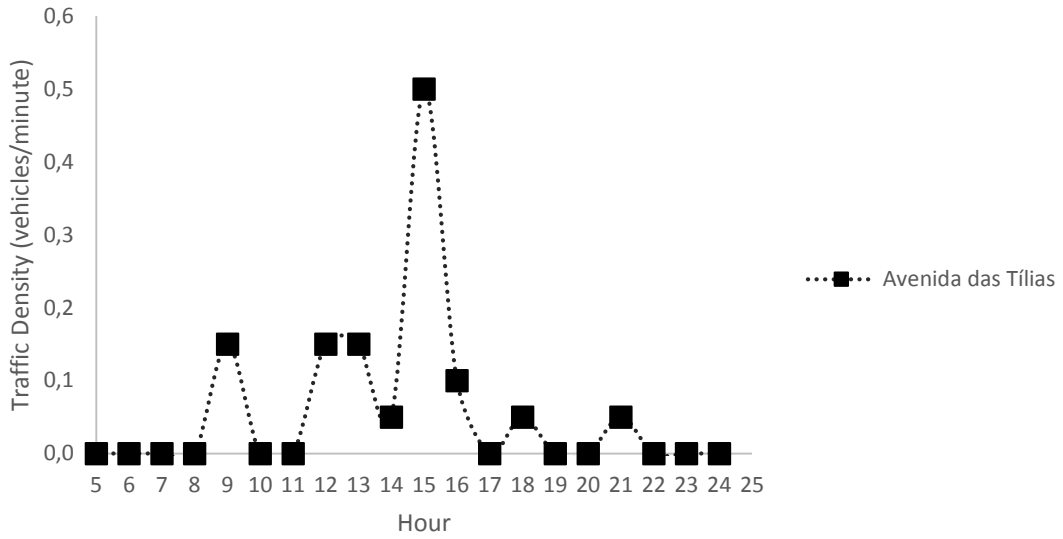


Figure 3.1.7- Average traffic density profile for the street surrounding H4.

As we can observe in Figure 3.1.7, overall traffic density is very low compared to the others homes. This is due to the fact that that this was a remote site. In addition, Tílias Av where RH1 was situated only gives access to local residents, at the end of the avenue there is local forest. Thus traffic in this street results only from cars of the few residents of Tílias Av.

3.1.5 Overview of the important properties of each home

The following Table 3.1.4 describes relevant comparisons of all homes of this study.

Table 3.1.4 - Relevant characteristics of the selected homes.

Properties		***H1***	***H2***	***H3***	***H4***
Building properties	Year of construction	1974	1994	1981	2000
	Main building materials	Concrete, wood, glass, metal	Concrete, wood, glass, metal	Concrete, wood, glass, metal	Concrete, wood, glass, metal.
	Number of floors	5	3	5	2

Number of occupants	Infants	2	1	1	1
	Adults	2	2	3	2
Location	Environment	Urban	Urban	Urban	Rural
Smokers		None	None	None	2
Heating systems		Non-existent	Fireplace (never used during campaign)	Fireplace (never used during campaign)	AVAC system
Pets		None	None	None	None
Average traffic density of nearby streets (vehicle min^{-1})		2.1	3.6	-	0.1

"-" – Not available.

3.2 Micro-environments

In all homes, the sampling campaign was performed in the living-rooms. In order to better understand the fate and behavior of indoor particles, these micro-environments are described in Table 3.2.1.

Table 3.2.1 - Main properties of sampling sites in four homes.

Micro-Environment	***Properties***	***H1***	***H2***	***H3***	***H4***
Living Room	Air volume (m^3)	66.1	117.3	116.1	146.9
	Surface area (m^2)[a]	109.1	171.1	-	203.4

[a] Total surface area in contact with room indoor air including walls, floor and ceiling.

	Main construction materials	Wood; concrete, paint, glass	Wood; concrete, paint, glass	Wood; concrete, paint, glass	Wood; concrete, paint, glass
	Ventilation	Natural	Natural	Natural	AVAC system
	Average temperature (º C)	21.6	21.7	22.3	21.6
	Average relative humidity (%)	61.6	54.7	60.4	59.2

"-" – Not available.

3.3 Sample Collection

The sampling period consisted in a total of 38 days of 2014, from April 8 to May 22, during both weekdays and weekends (and including national holidays). The specific periods for each home are described in Table 3.3.1.

Table 3.3.1 - Sampling period for each home.

Home	***H1***	***H2***	***H3***	***H4***
Sampling Period	April 8 – 16	April 16 – 22; April 24 – 28	May 2 – 10	May 15 – 22

Two instruments P-Trak (TSI, Inc., USA, Model 8525; (Figure 3.3.2) were used to continually sample indoor and outdoor UFP number concentration. Sampling of PM_1, $PM_{2.5}$ and PM_{10} was made indoor by Dust Trak DRX (TSI, Inc., USA, Model 8533) that operated continuously indoors (Figure 3.3.1).

In order to prevent P-Trak instrument sampler of malfunctioning and misleading particle number concentration measurements, it was necessary to immerse its cylinder with isopropyl alcohol in periods no bigger that 4-5 hour long, continuously during the campaigns in all homes.

Figure 3.3.2 - Used TSI P-Trak Model 8525.

Figure 3.3.1 - Used TSI Dust Trak DRX 8533.

In addition, gravimetric measurements of PM were performed both in indoor and outdoor air of homes using personal modular impactor for $PM_{2.5}$ (SKC, Inc., USA; Figure 3.3.4). Quartz fiber filters (ø 25 and 37 mm filter; SKC, Inc., USA) were used to collect coarse (i.e. >2.5µm;) and fine particles ($\leq$ 2.5µm). Four universal air sampling pumps (SKC, Inc., USA; models 224-PCTX8 and AirChek XR5000) were used with air flow of 3 L min^{-1}. Flow calibration of thes pumps was conducted daily by primary flow meter (Dry Cal DC- Lite, BIOS International Corporation, USA; model DCL-ML). Two flexible tubes were used to connect the pumps to the impactors, as well as for calibration (Tygon Tube, Saint-Gobain Corporation S.A., France).

Indoor temperature and relative humidity were continuously measured by Mini data logger was used (Testo, Germany, model 174H) with logging interval of 1 min.

Figure 3.3.4 - Personal modular impactor for $PM_{2.5}$

Figure 3.3.3 - Mini data logger Testo 174H

Further sampling specifications for each instruments are present in the following Table 3.3.2.

Table 3.3.2- Sampling details for different equipment's PM levels sampling.

Equipment	***Log interval (min)***	***Daily Logging Period***	***Sampling Height (m)***
P-Trak	1	All day	1 to 1.5
Dust Trak	1	All day	1 to 1.5
Testo 174H	1	All day	1 to 1.5

A digital laser range finder was used (Bosch, Germany, model PLR 50) for the measurements of H1 dimensions, in order the draw an estimated blueprint of the house. For the elaboration of H2 and H4 blueprint, the original blue prints of the homes were used. All drawing were made in AutoCAD software.

All available blueprints are present in Appendix A.

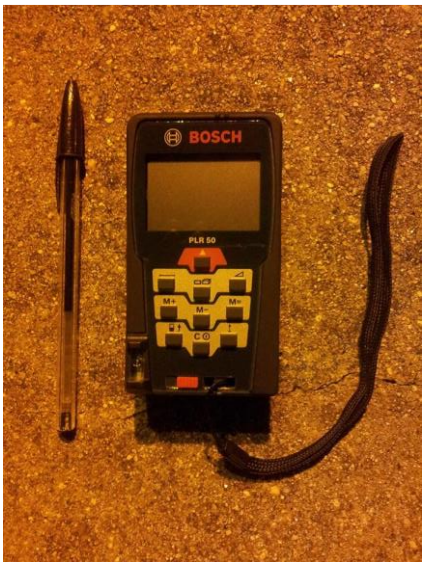


Figure 3.3.5 - Digital laser range finder Bosch PLR 50.

3.4 Indoor Sources and Activity Patterns

In every home, the occupants filled daily detailed questionnaires. One questionnaire was dedicated to registering potential sources of UFP and PM, where the occupants marked time when these activities/sources were conducted/used, in order to cross-reference them with concentration levels. The second questionnaire was dedicated to the occupancy/activities of room where sampling equipment was placed. The last questionnaire focused on schedule of children's activities and their physical activity during the sampling. The examples of the questionnaires are in Appendix B.

3.5 Dose Rate

In order to quantify the level of exposure for children and adults in the studied homes, the dose rate was calculated according to the following Equation 1:

$$Dose\ Rate\ (D) = \left(\frac{BR}{BW}\right) \times C_{WA} \times OF \times N\ (1)$$

In which:

- D is the age-specific dose rate (particle number $kg^{-1}d^{-1}$);
- BW is age-specific body weight (kg);
- C_{WA} is the age-specific weighted average concentration (particle number l^{-1});

- OF is the occupancy factor (considered always one);
- N is the total time spent by children in the home (min d^{-1}).

Age-specific breathing rates and body weights were extracted from the available literature, and the relevant values are presented in Table 3.5.1.

Table 3.5.1 - Age-specific body weight values, extracted from U.S. EPA (2011).

Age Group	***Mean (kg)***
3 to < 6 years	18.6
21 to < 65 years	76.0

Table 3.5.2 -Age and activity level specific breathing rates, extracted from U. S. EPA (2011).

Age Group	***Activity Level***	***Mean (l min^{-1})***
3 to < 6	Sleep or Nap	4.3
	Sedentary / Passive	4.5
	Light Intensity	11.0
	Moderate Intensity	21.0
	High Intensity	37.0
21 to < 61	Sleep or Nap	4.8
	Sedentary / Passive	4.6
	Light Intensity	12.5
	Moderate Intensity	27.5
	High Intensity	51.0

The age-specific breathing rate varied during the time children spend at homes with the type of activity intensity of the child. Therefore, in order to obtain values that are more liable for the Breathing Rate, its weighted average was calculated according to the following Equation 2:

$$BR_{WA} = \Sigma \frac{BR_i \times n_i}{N} \quad (2)$$

Where:

- BR_{WA} is the age-specific weighted average concentration (l min^{-1});
- BR_i is the average concentration in a specific location (l min^{-1});
- n_i is the number of hours spent by age-specific children in that location (h);
- N is the total number of hours spent by age-specific children in the home (h).

The following Equation 3 was considered for the calculation of the age-specific values of C_{WA}:

$$C_{WA} = \Sigma \frac{C_i \times n_i}{N} \quad (3)$$

Where:

- C_{WA} is the age-specific weighted average concentration (particle number l^{-1});
- C_i is the average concentration in a specific location (particle number l^{-1});
- n_i is the number of hours spent by age-specific children in that location (h);
- N is the total number of hours spent by age-specific children in the home (h).

3.6 Statistical Analysis

For the statistical analysis of the data from these campaigns, a T- test was performed ($P < 0.05$; two tailed) in order to study the significant of the sample from different sites and the existing differences between calculated averages.

3.7 References

INE. 2012. *Censos 2011 - Instituto Nacional de Estatística* [Online]. Available: http://www.ine.pt/xportal/xmain?xpid=INE&xpgid=ine_base_dados&contexto=bd&selTab=tab2 [Accessed 01/06/2014 2014].

U.S. EPA, 2011. Exposure Factors Handbook 2011 Edition (Final). *U.S Environmental Protection Agency.* Washington, DC, U.S. Environmental Protection Agency

4. Results and Discussion

4.1 Particle Concentrations and Comparison with other Studies

In this section, the obtained results for indoor concentrations of the studied pollutants are presented as well as a comparison with other international studies.

4.1.1 Ultrafine Particles

Mean number concentrations of ultrafine particles in the selected homes are presented in Table 4.1.1. These results consider particle number concentrations of respective analyzed indoor environments, namely the concentration of the living/dining rooms of each household environment.

Table 4.1.1 - Ultrafine number concentration: mean and ranges at four homes (particle cm^{-3})

	Mean	***Range*** *(min – max)*
H1	1.24×10^4	2.4×10^3 - 2.1×10^5
H2	1.09×10^4	1.0×10^3 - 1.0×10^5
H3	1.11×10^4	1.3×10^3 - 8.8×10^4
H4	1.62×10^4	2.2×10^3 - 2.0×10^5

The comparison of UFP mean at H1, H2 and H3 showed that the levels were not significantly different ($P < 0.05$).

The statistical analysis of the results obtained in this study indicated that number concentrations were significantly lower ($P < 0.05$) in non-smokers homes (H1, H2 and H3) than in H4.

H4 presents a mean value 1.5 times higher than H2. As for the comparison between H3 and H4, it reveals that H4 has a mean value 1.5 times higher than H3. Concerning H1 and H4, H4 presents a mean value 1.31 times higher than H1. Similar ranges were due to the smaller volume of room H1 in comparison with H4, leading to accumulation of pollutants and thus to higher peak values regarding H4 and at the same time lower residence times (compared to H4) due to the absence of a smoking source, which perpetuates higher concentrations longer in time. As demonstrated, UFP mean concentration values were much lower in non-smokers residences than in homes occupied by smokers (i.e. H4).

Comparison between non-smokers homes may bring further understanding of particle's dynamics in these indoor environments. H1 presents a 1.13 times higher mean value than H2. Comparing H1 and H3, H1 reveals to have an average 1.11 times higher than H3. It is possible to infer about the relevance of the confinement parameter for indoor particles, since it is determinant for the dispersion of these pollutants. As for H3 and H2, the mean values showed to be similar, with H3 presenting 1.01 times higher average values than in H2. These values can be justified by the similar volumes of the two spaces in which the sampling took place in each home (117.28 m^3 for H2 and 116.10 m^3 for H3 – see Appendix A.2 and. A.3 for details. The frequency and duration of UFP producing activities influenced very probably these results, since at H2 presents much higher number of the respective activities than H3, as can be further observed in Section 4.3.

Table 4.1.2 provides information about levels from UFP from other studies in order to compare the obtained ranges of concentrations.

Table 4.1.2 - Comparison between the present study and other international studies on indoor UFP

Country	***Fraction Particle***	***Mean***	***Study conditions***	***Reference***
Portugal	UFP	1.24×10^4 particle cm^{-3} (H1) 1.09×10^4 particle cm^{-3} (H2) 1.11×10^4 particle cm^{-3} (H3) 1.62×10^4 particle cm^{-3} (H4)		This study
Canada	UFP	9.7×10^3 particle cm^{-3} [2] 9.3×10^3 particle cm^{-3} [3]	50 homes sampled each season; study carried out in winter and summer seasons of 2010; 24h measurements for 7 days.	(Kearney et al., 2014)
Germany	20 – 1000 nm[4]	9.00×10^3 particle cm^{-3} NR[5] 6.00×10^3 particle cm^{-3} BMDR[6]	192 residences [7]; measurements made for 8h during winter of 2007;	(Fittschen et al., 2013)
Denmark	10 – 300 nm[4]	2.91×10^4 particle cm^{-3}	56 residences (non-smokers) ; measured continuously for a 45h period;	(Bekö et al., 2013)
China	UFP	1.64×10^4 particle cm^{-3}	4 homes were monitored for 48-71h between June and August 2009;	(Mullen et al., 2011)
Canada	UFP	7.70×10^3 particle cm^{-3} [8] 9.65×10^3 particle cm^{-3} [11] 7.88×10^3 particle cm^{-3} [9]	48 and 45 homes (non-smokers) in winter and summer respectively;	(Wheeler et al., 2011)
Canada	20 – 100 nm	7.99×10^3 particle cm^{-3} [10] 1.03×10^4 particle cm^{-3} [11]	45 homes (non-smokers) in summer 2005 (July-August); 47 homes in the 2006 winter (asthmatic children); 45 homes in the summer of 2006; all measurements over 5 consecutive days for 24h (sample for 10 min of each hour).	(Kearney et al., 2011)
USA	UFP	1.70×10^4 particle cm^{-3}	7 houses; Studied during 2007-2009	(Bhangar et al., 2011)

[2] In winter
[3] In summer
[4] Fine Mode: Nuclei (UFP) and Accumulation Mode
[5] Normal Residences
[6] "Black Magic Dust" Residences
[7] 137 Normal Residences and 55 "Black Magic Dust" residences. The referred study is dedicated to fine and ultrafine particles characterization in private residences also regarding to a phenomenon of enhanced soiling, known as "black magic dust" staining in walls and ceilings, which is supposedly correlated with SVOC's emissions.
[8] Summer 2005
[9] Summer 2006
[10] Summer 2005 and Summer 2006
[11] Winter 2006

Greece	10 – 400 nm[12]	Warm period: 1.40×10^4 particle cm^{-3} [13] 2.00×10^4 particle cm^{-3} [14] 2.60×10^4 particle cm^{-3} [15] Cold period: 2.30×10^4 particle cm^{-3} [13] 1.30×10^4 particle cm^{-3} [14] 3.40×10^4 particle cm^{-3} [15]	3 homes; sampling was performed in 2 campaigns during cold period (March-April) and warm period (November-December) Each home for a period of 1-2 weeks;	(Diapouli et al., 2011)
	100 – 3000 nm[12]	Warm period: 1.50×10^3 particle cm^{-3} [13] 1.60×10^3 particle cm^{-3}[14] 3.10×10^3 particle cm^{-3} [15] Cold period: 1.80×10^3 particle cm^{-3}[13,15] 2.00×10^3 particle cm^{-3} [14]		
USA	UFP	4.90×10^4 particle cm^{-3} [13] 5.30×10^4 particle cm^{-3}[14] 6.20×10^4 particle cm^{-3} [15] 1.90×10^4 particle cm^{-3} [16] 4.50×10^4 particle cm^{-3} [17]	5 residences; sampling was conducted for 1 day for each home during weekday (mid-afternoon to evening) during January 2006;	(McAuley et al., 2010)
USA	10 – 50 nm[18] 50 – 100 nm[18] 10 – 100 nm[18] 100 – 500 nm[19] 500 – 1000 nm[19]	5.25×10^3 particle cm^{-3} 2.02×10^3 particle cm^{-3} 7.27×10^3 particle cm^{-3} 8.01×10^2 particle cm^{-3} 1.94 particle cm^{-3}	1 home; sampling conducted from November 1999 to March 2000;	(Ogulei et al., 2006)
Australia	UFP	1.24×10^4 particle cm^{-3} [20] 1.82×10^4 particle cm^{-3} [21]	15 houses; continuous monitoring for more than 48h during winter of 1999;	(Morawska et al., 2003)
USA	UFP	1.87×10^4 particle cm^{-3}	1 home measured continuously for 18 months;	(Wallace and Howard-Reed, 2002)

[12] Fine Mode: Nuclei (UFP) and Accumulation Mode
[13] Residence 1
[14] Residence 2
[15] Residence 3
[16] Residence 4
[17] Residence 5
[18] Fine Mode: Nuclei Mode (UFP)
[19] Fine Mode: Accumulation mode
[20] Under non-activity conditions
[21] During indoor activity

Comparing the overall obtained results with existing international studies it is possible to conclude that USA and China presents higher values for indoor UFP concentrations. On the other hand, in Canada, Denmark and Germany show lower levels were reported, while Greece and Australia showed UFP levels mostly similar to Portugal.

4.1.2 Particulate Matter

Table 4.1.3 presents the means and ranges of indoor PM in the four homes.

Table 4.1.3 - Particulate Matter concentrations in all household environments (μg m^{-3}).

	PM₁		***PM₂.₅***		***PM₁₀***	
	Mean	*Range (min – max)*	*Mean*	*Range (min – max)*	*Mean*	*Range (min – max)*
H1	4.59×10^1	1.60×10^1 - 1.23×10^3	4.76×10^1	1.60×10^1 - 1.25×10^2	5.13×10^1	1.60×10^1 - 1.52×10^2
H2	2.86×10^1	4.00 - 2.39×10^3	2.99×10^1	5.00 - 2.49×10^2	3.27×10^1	5.00 - 8.89×10^2
H3	1.81×10^1	4.00 - 8.50×10^1	1.93×10^1	5.00 - 9.50×10^1	2.16×10^1	5.00 - 9.70×10^1
H4	1.34×10^2	1.00×10^1 - 9.78×10^2	1.36×10^2	1.00×10^2 - 9.96×10^3	1.40×10^2	1.30×10^1 - 1.01×10^3

As expected H4 that was occupied with 2 smokers presents higher mean values for all PM fractions compared with the remaining homes under study.

In fact, for PM_1 the mean values obtained in H4 were 7.4 times higher than H3. For $PM_{2.5,}$ H4 presented mean of 7.0 times higher than in H3. As for PM_{10} fraction, the values obtained in H4 were 6.48 times higher for the comparisons of mean values with H3.

In relation to H2 in PM_1 values, H4 presented a 4.7 times higher value for the mean average. For $PM_{2.5}$ fraction, H4 presented mean values 2.9 times higher than H2. PM_{10} fraction showed also higher values in H4 than in H2 with a proportion of 4.3 between the two mean values.

Finally, PM_1 in H4 demonstrated to have a mean average value 2.9 times higher than H1. For $PM_{2.5,}$ H4 presented mean values 2.8 times higher than H1. Regarding PM_{10}, H4 had 2.7 times higher mean values.

Overall, it is possible to conclude that for all PM fractions in all homes where the occupants were non-smokers, the highest mean values were obtained for H1, similarly as to UFP values.

For PM_1, H1 presented mean values 2.5 times higher than H3. For $PM_{2.5}$ H1 presents 2.5 times higher mean values than H3. For PM_{10} faction, the mean obtained values are 2.4 higher than in H3.

In comparison to H2, H1 revealed mean values for PM_1 fraction 1.60 higher than H2. For $PM_{2.5}$, this comparison was 1.59 times higher than H2 for mean obtained. Regarding PM_{10}, H1 presented mean values 1.57 times higher than at H2. H1 confinement compared with H2 and H3 may justify this occurrence, since as we can see further in section 4.3, H2 presented higher number of sources than H1.

Site H3 presented the lowest values for all PM fractions from four homes. In relation to H2, H3 presented mean values for PM_1 1.6 times lower. For $PM_{2.5}$, H3 showed a mean value 1.6 times lower. Regarding PM_{10}, mean value obtained in H3 was 1.5 times lower than in H3.

From its definition, PM fraction aggregates a big group of aerodynamic diameters. For example, PM_{10} fraction refers to particles with 10 μm and smaller. In this way, in order to evaluate the contribution of smaller fractions to coarser PM, their ratio was calculated. The results are presented in Table 4.1.4.

Table 4.1.4 – Ratio between different PM fractions.

	$PM_{2.5}/PM_{10}$		***PM_1/PM_{10}***		***$PM_1/PM_{2.5}$***	
	Mean	*Range (min-max)*	*Mean*	*Range (min-max)*	*Mean*	*Range (min-max)*
H1	0.932	0.566 - 1.00	0.897	0.526 - 1.00	0.962	0.855 - 1.00
H2	0.923	0.423 - 1.00	0.881	0.406 - 1. 00	0.955	0.727 - 1.00
H3	0.897	0.625 - 1.00	0.841	0.568 - 1.00	0.938	0.769 - 1.00
H4	0.944	0.460 - 1.00	0.921	0.426 - 1.00	0.974	0.838 - 1.00

In order to better understand the obtained results of this study, the following Table 4.1.5 presents a comparison between different studies on indoor PM concentrations from different international assessments.

Table 4.1.5 - Comparison between the present study and other international studies on indoor PM.

Country	***Fraction Particle***	***Mean***	***Note***	***Reference***
Portugal	PM_1	45.9 µg m^{-3} H1 28.6 µg m^{-3} H2 18.1 µg m^{-3} H3 134 µg m^{-3} H4		This study
	$PM_{2.5}$	47.6 µg m^{-3} H1 29.9 µg m^{-3} H2 19.3 µg m^{-3} H3 136 µg m^{-3} H4		
	PM_{10}	51.3 µg m^{-3} H1 32.7 µg m^{-3} H2 21.6 µg m^{-3} H3 140 µg m^{-3} H4		
Lithuania	PM_1	3.1 µg m^{-3} [22] 14.7 µg m^{-3} [23]	50 apartments; measurements made during 1 week (Monday to Friday) during the two heating seasons of 2011 and 2012	(Prasauskas et al., 2014)
	$PM_{2,5}$	4.5 µg m^{-3} [22] 18.1 µg m^{-3} [23]		
	PM_{10}	13.6 µg m^{-3} [22] 38.4 µg m^{-3} [23]		
Germany	PM_{10}	58 µg m^{-3} [24] 73 µg m^{-3} BMDR[26]	192 residences[25]; measurements made for 8h during winter of 2007	(Fittschen et al., 2013)
	$PM_{2.5}$	15 µg m^{-3} µg/m^3 [24] 27 µg m^{-3} BMDR [26]		

[22] Minimum mean value
[23] Maximum mean value
[24] Normal Residences
[25] 137 Normal Residences and 55 "Black Magic Dust" residences. The referred study is dedicated to fine and ultrafine particles characterization in private residences also regarding to a phenomenon of enhanced soiling, known as "back magic dust" staining in walls and ceilings, which is supposedly correlated with SVOC's emissions.
[26] "Black Magic Dust" Residences

U.S.A.	≥ 300 nm[27]	97.2 ± 2.3 particle/cm^3	116 residences[28] ; conducted from January to December 2006; homes sampled a single time;	(Leavey et al., 2012)
Canada	$PM_{2.5}$	7.9 µg m^{-3} [29] 10.2 µg m^{-3} [30] 8.0 µg m^{-3} [31] 8.4 µg m^{-3} [32]	48 and 45 homes (non-smokers) in winter and summer respectively;	(Wheeler et al., 2011)
Italy	PM_1	29.6 µg m^{-3}	60 homes; sampling during summer (May to September 2007) and winter (March 2008) for ≈ 24h;	(Cattaneo et al., 2011)
	$PM_{2.5}$	33.2 µg m^{-3}		
	PM_{10}	40.9 µg m^{-3}		
Greece	PM_{10}	Warm period: 27.1 µg m^{-3} [13] 30.8 µg m^{-3} [14] 47.3 µg m^{-3}[15] Cold period: 30.0 µg m^{-3} [13] 25.1 µg m^{-3} [14] 40.4 µg m^{-3} [15]	3 homes; sampling was performed in 2 campaigns during cold period (March-April) and warm period (November-December) Each home for a period of 1-2 weeks;	(Diapouli et al., 2011)
Canada	$PM_{2.5}$	Winter: 9.42 µg m^{-3} Summer: 9.51 µg m^{-3}	106 and 111 homes under study in winter and summer $PM_{2.5}$ measured over 5 days.	(Héroux et al., 2010)
Bangladesh	PM_{10}	178 µg m^{-3} 332 µg m^{-3} 104 µg m^{-3} 159 µg m^{-3} 188 µg m^{-3}	5 rural homes; conducted from February to March 2006, 4h sampling;	(Begum et al., 2009)
USA	$PM_{2.5}$	40.3 µg m^{-3}	150 homes; monitoring in 1st, 3rd and 6th month, continuously for 3 days.	(McCormack et al., 2009)
	$PM_{2.5-10}$	17.4 µg m^{-3}		
China	$PM_{2.5}$	45.0 µg m^{-3}	34 homes; sampling from October 1999 to March 2000 (fall and winter in Hong Kong);	(Chao and Wong, 2002)
	PM_{10}	63.3 µg m^{-3}		

[27] Accumulation and coarse mode
[28] Asthmatic children below 7 years old
[29] Winter 2005
[30] Summer 2005
[31] Winter 2006
[32] Summer 2006

Comparing this study with the above international ones referred above (Table 4.1.5), it can be concluded that Canada shows lower PM levels in homes than the ones obtained in this study. As expected, Bangladesh and China (PM_{10}) showed much higher levels for indoor particulate matter. Study from Lithuania presented generally lower levels for PM_1 and $PM_{2.5}$ and similar concentration ranges of PM_{10}. The study from Germany showed similar values regarding $PM_{2.5}$. Italian and Greek studies also show identical results on PM_1, $PM_{2.5}$ and PM_{10}. Finally, the studies from USA and China ($PM_{2.5}$) show also similar ranges of PM levels in indoor air of homes.

Analyzing the ratios between the different PM fractions in all homes, it can be concluded that in general, PM1 was the main constituent of PM_{10}. In fact, Figures 4.1.1- 4.1.4 show that PM_1 represents the highest ratio of PM_{10} in all homes (0.897 to H1, 0.881 to H2, 0.841 to H3 and 0.921 to H4). Consequently $PM_{1\text{-}2.5}$ (i.e. particles with aerodynamic diameter bigger than 1 µm and smaller than 2.5 µm) contributed 0.035 in H1, 0.041 in H2, 0.103 in H3 and 0.023 in H4 to the PM_{10} concentration. Finally, particles with an aerodynamic diameter bigger than 2.5 µm and smaller than 10 µm ($PM_{2.5\text{-}10}$) accounted for 0.068 of PM_{10} in H1, 0.077 in H2, 0.055 in H3 and 0.056 in H4.

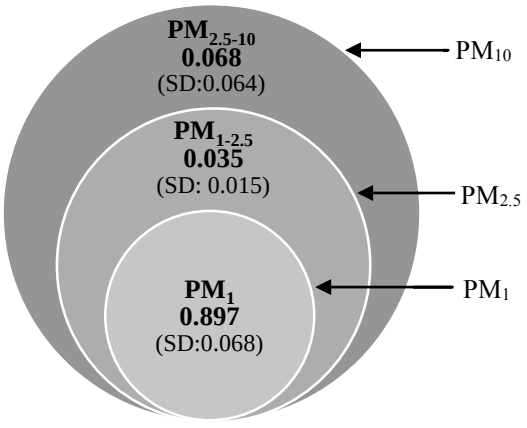


Figure 4.1.1 - Graphical representation of fraction contribution for PM_{10} in H1.

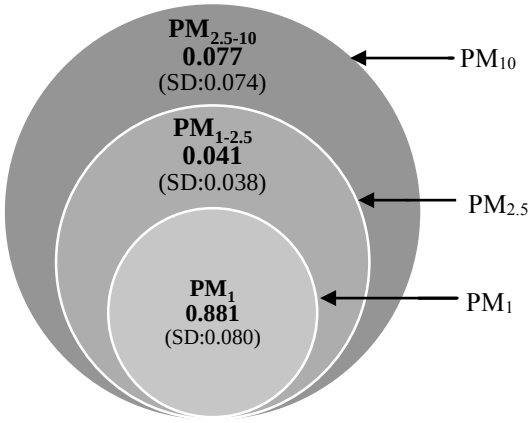


Figure 4.1.2 - Graphical representation of fraction contribution for PM_{10} in H2.

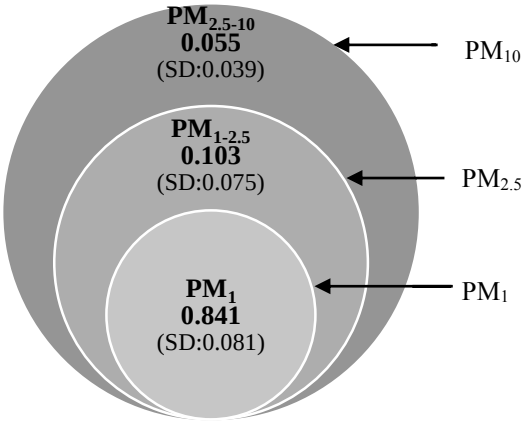


Figure 4.1.3 -Graphical representation of fraction contribution for PM_{10} in H3.

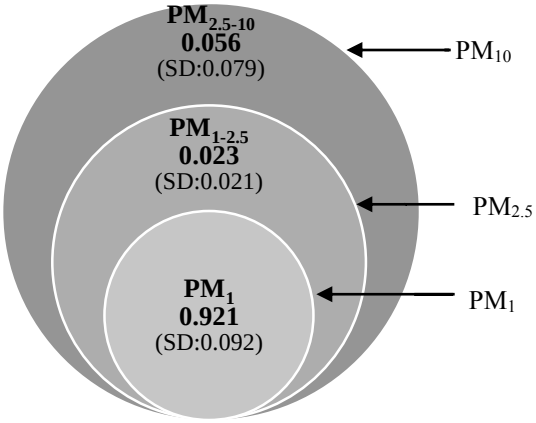


Figure 4.1.4 - Graphical representation of fraction contribution for PM_{10} in H4.

4.2 Indoor and Outdoor Concentration

4.2.1 Ultrafine Particles

In order to compare indoor and outdoor levels of pollution, measurements were performed concurrently outdoors of all homes These measurements allow a better understanding of indoor concentration and particle behavior in terms of penetration from outdoor sources and transportation. The comparison of indoor and outdoor values are presented in Table 4.2.1 Aiming to analyze the influence of outdoor sources in indoor UFP concentrations, the indoor/outdoor (I/O) ratio was calculated.

Table 4.2.1-Indoor and outdoor UFP levels and outdoor contribution (particle number cm^{-3}).

	Indoor		***Outdoor***		***I/O***	
	Mean	*Range (min-max)*	*Mean*	*Range (min-max)*	*Mean*	*Range (min-max)*
H1	1.24×10^4	$2.36 \times 10^3 – 2.08 \times 10^5$	1.24×10^4	$3.75 \times 10^3 – 5.22 \times 10^4$	1.02	0.24 – 28.26
H2	1.09×10^4	$1.04 \times 10^3 – 1.03 \times 10^5$	8.05×10^3	$5.40 \times 10^2 – 1.03 \times 10^5$	1.88	0.06 – 48.14
H3	1.11×10^4	$1.27 \times 10^3 – 8.82 \times 10^4$	1.05×10^4	$1.07 \times 10^3 – 8.15 \times 10^4$	1.25	0.09 – 17.06
H4	1.62×10^4	$2.27 \times 10^3 – 2.03 \times 10^5$	7.78×10^3	$2.13 \times 10^2 – 6.14 \times 10^4$	2.56	0.08 – 84.70

H4 was located in a rural place, with the lower traffic density of all sampling locations. This home shows the lowest level of outdoor UFP concentrations. In fact, H4 shows an average outdoor value of 1.6 times lower than in H1. H1 was located on the 4th floor of a multiunit building whereas sampling in H4 took place in a height corresponding to 1st floor. As the main

source of outdoor UFP (i.e. vehicle traffic) is located on the ground level, dispersion of UFP might have a significant influence on the sampled levels. H3 presents a mean value 1.4 times higher than H4. Regarding H2, the mean value obtained is 1.0 times higher than H4. The lower difference to H4 regarding the difference of H2 and H3 can be justified by the geographical layout of this area compared with the other two homes. Custóias is designated as a "sub-urban zone" with overall more intense traffic density and lower construction density of the surrounding buildings, thus allowing higher dispersion of pollutants in outdoor air.

Comparing the results from non-rural places, this fact becomes evident. In fact, H3 mean value is 1.2 times higher than H2. H1 presents a mean value 1.5 times higher than in H2. The sampling of H1 and H3 took place on a 4th floor, whereas outdoor sampling in H2 was performed in ground floor. H1 presented a mean value 1.2 times higher than the mean value obtained in H3.

In order to further analyse the influence of outdoor concentrations in indoor UFP levels the indoor/outdoor ratios (I/O) were calculated.

- If I/O < 1 – Indoor concentration values were probably mostly affected by outdoor sources through transportation and infiltration mechanisms (i.e. outdoor air was the predominant source of UFP indoors);
- If I/O > 1 – Indoor concentration values were mostly due to indoor UFP producing activities (i.e. indoor sources were the predominant contributor to indoor UFP).

In all homes, I/O ratio was bigger than 1. Indoor sources produce high UFP number concentrations that are aggravated by the confined space inside residences, not enabling the dispersion of these pollutants easily. The highest ratio is observed in H4 where the contribution of indoor sources was higher due to the fact that the occupants smoked. H1 presents the lowest I/O ratio, leading to the conclusion that overall indoor and outdoor concentrations were similar.

Figure 4.2.1 presents the means and the statistical parameters (minimum, 25th percentile, median, 75th percentile and maximum values) UFP number concentrations in both indoor and outdoor air of four homes. It is also important to enhance that in order to better perceive the obtained set of values, the y-axis has a discontinuous scale (between 100,000 and 200,000).

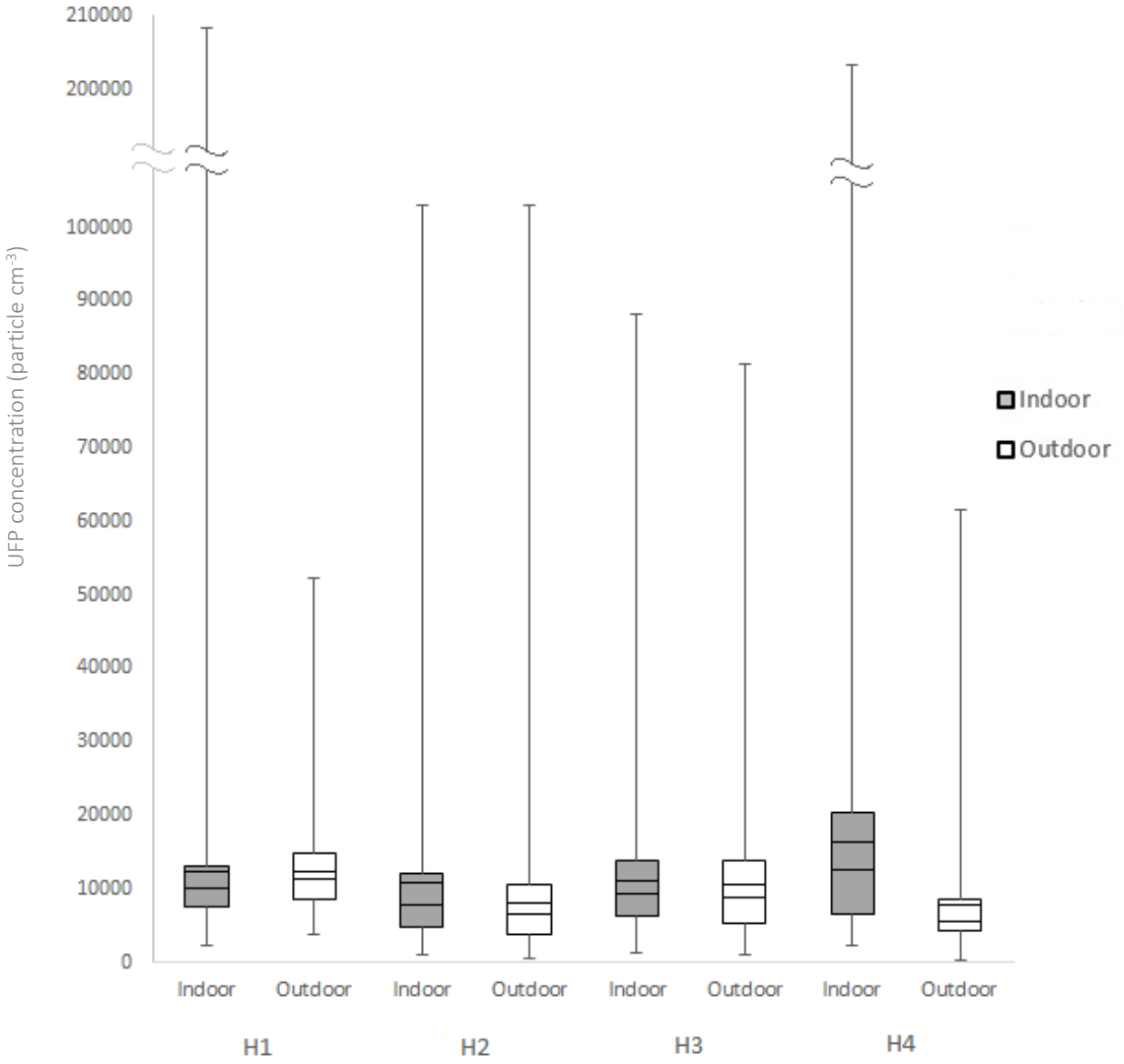


Figure 4.2.1 - Ultrafine particle concentrations at four homes: maximum, 75th percentile, average, median, 25th percentile, and minimum (particle cm^{-3}).

4.2.2 Particulate Matter

In order to compare indoor and outdoor $PM_{2.5}$ mass concentrations, these parameters were calculated by gravimetric method. The obtained results are presented in Table 4.2.2.

Table 4.2.2 - Indoor and outdoor $PM_{2.5}$ by gravimetric method (μg m^{-3}).

	Indoor		***Outdoor***		***I/O***	
	Mean	*Range (min-max)*	*Mean*	*Range (min-max)*	*Mean*	*Range (min-max)*
H1	3.92×10^1	1.26×10^1 - 6.09×10^1	4.76×10^1	3.10×10^1 - 1.03×10^2	8.97×10^{-1}	2.96×10^{-1} – 1.27
H2	2.99×10^1	1.44×10^1 - 5.20×10^1	2.08×10^1	1.05×10^1 - 2.91×10^1	1.47	1.02 – 1.90
H3	1.69×10^1	4.64 - 2.72×10^1	7.09	2.32 - 1.22×10^1	3.47	1.10 – 1.00×10^1
H4	1.15×10^2	8.88×10^1 - 1.38×10^2	7.76×10^1	7.05×10^1 - 8.69×10^1	1.50	1.13 – 1.96

H4 presents an I/O ratio higher than 1, indicating that indoor $PM_{2.5}$ concentrations originated mostly from indoor sources; H4 was a smoking home. In H1 the ratio was lower than 1 suggesting that indoor $PM_{2.5}$ resulted probably from outdoor contribution. In H2 and H3, indoor sources were on average the main contributors for overall indoor $PM_{2.5}$ levels. However, high maxima values of ratio (especially at H3) indicates the occurrence and some contribution from indoor sources.

4.3 Evaluation of Ultrafine Particle Sources in Indoor Environment

As demonstrated in section 4.2.1., the UFP concentrations in all four homes were strongly influenced by indoor sources and activities. These sources and activities are fully dependent on the home characteristic but also on occupant's habits of living and behavior inside the home. In order to better understand the different dynamics between homes, a detailed count of UFP producing events in each house was made from the existent questionnaires. The absolute increase of UFP levels during each activity was also registered. These number results are presented in Table 4.3.1 as well as the ranges observed of the increases.

Table 4.3.1- Number of UFP producing events and respective detected range increase in UFP concentrations (particle cm^{-3}) in four homes.

		H1	***H2***	***H3***	***H4***	***Total***
Cooking	Soup	3 (3.82×10^3 - 5.74×10^4)	2 (8.74×10^3)[gg]	2 (2.63×10^3)[gg]	1 (7.95×10^3)	8
	Boiling	11 (3.78×10^2 - 1.62×10^5)	10 (4.82×10^2 - 1.18×10^4)	1 [hh]	4 (3.49×10^3 - 5.48×10^3)	26
	Stew	0	7 (1.18×10^4 - 5.56×10^4)	0	0	7
	Frying	0	7 (7.99×10^3 - 5.53×10^4)	4 (1.04×10^4 - 1.80×10^4)	10 (2.77×10^3 - 1.17×10^5)	21
	Baking	0	8 (8.43×10^2 - 5.08×10^4)	2 (3.64×10^2 - 6.24×10^3)	0	10
	Grilling	1 (3.21×10^4)	0	0	0	1
Use of electric toaster		4 (5.74×10^4 - 1.86×10^5)	3 (1.38×10^4 to 5.31×10^4)	7 (2.95×10^3 to 7.64×10^4)	0	14
Use of oven		8 (electric) (4.74×10^4- 1.93×10^5)	5 (gas) (1.81×10^4 - 5.38×10^4)	1 (electric)[hh]	2 (electric) (1.01×10^5 - 2.00×10^5)	16
Candle/incense burning		0	1 (1.93×10^4)	0	0	1
Smoking		0	0	0	60 (1.34×10^3 - 5.73×10^4)	60
Hair spaying		0	1 (1.09×10^4)	0	0	1
Use of cleaning products		1 (No effect)	9 (9.06×10^2 - 1.86×10^4)	1 (5.01×10^3)	0	11
Use of furnish polish		0	1 (No effect)	2 (2.92×10^3)	0	3

[gg] This event only occurred once independently from other UFP producing events.

[hh] The only time this event occurred was together with other source event

Ironing	1 (7.68×10^{3})	1 (No effect)	3 (1.00×10^{4} - 2.25×10^{4})	0	5
Ventilation	3 [(-1.92×10^{4}) - 4.46×10^{3}]	4 [(-2.02×10^{4}) - 7.46×10^{2}]	0	12 [(-1.92×10^{4}) – (-4.07×10^{2})]	19
Total	32	59	23	89	**203**

There was observed a total number of 203 UFP producing events in all the homes. Overall, for the same type of activities, the values of the UFP increase obtained for the different homes were coherent.

UFP increases due to cooking activities were observed in a total number of 73 in all homes. Specifically, cooking soup was observed in a total of 8 times with an overall range of 2.63×10^{3} to 5.74×10^{4} particle cm^{-3}. The lowest increase was observed in H3 and the maximum level was observed in H1. Boiling was observed in a total number of 26 times in all homes and the increases varied from 3.78×10^{2} to 1.62×10^{5} particle cm^{-3} with both values registered in H1. Stewing occurred 7 times, always in H2 and the increases ranged from 1.18×10^{4} to 5.56×10^{4} particle cm^{-3}. Frying occurred in a total number of 21 times and the range varied from 2.77×10^{3} to 1.17×10^{5} particle cm^{-3}, with the both levels observed in H4. Baking was performed in a total number of 10 times and the UFP increase varied from 3.64×10^{2} to 5.08×10^{4} particle cm^{-3} with the lowest value observed in H3 and the highest value in H2. Grilling occurred only once in H1 and the detected increase in UFP concentration was of 3.21×10^{4} particle cm^{-3}.

The usage of electrical toaster occurred in a total number of 14 times and the UFP increases due to this activity ranged from 2.95×10^{3} to 1.86×10^{5} particle cm^{-3}. The lowest increase was observed in H3 and the maximum one in H1.

The usage of oven occurred in a total number of 16 times of which 5 of them occurred with gas oven and the remaining 11 with electric. The range of increase for electric stove is from 4.74×10^{4} (H1) to 2.00×10^{5} (H4) particle cm^{-3}. The range of increase for gas stove was from 1.81×10^{4} to 5.38×10^{4} particle cm^{-3} and both occurred in H2.

Incense burning was only observed once in H2 and resulted in an increase of 1.93×10^4 particle cm^{-3}.

Smoking occurred in a total of 60 times in H4 and the range of increases detected varied from 1.34×10^3 to 5.73×10^4 particle cm^{-3}.

Hair spraying only occurred once in H2 and resulted in an increase of 1.09×10^4 particle cm^{-3}.

The usage of cleaning products occurred in a total of 11 and the UFP concentration increased due to this activity varied from 9.06×10^2 to 1.06×10^4 particle cm^{-3}, with both values observed in H2.

The usage of furnish polish occurred 3 times, but only once this activity had an influence on indoor UFP levels resulting in an increase of 2.92×10^3 particle cm^{-3} observed in H3.

Ironing occurred 5 times during the sampling in residences and the peak increase concentration observed ranged from 7.68×10^3 to 2.25×10^4 particle cm^{-3} observed in H1 and H3 respectively.

Ventilation occurred 19 times, taking into account all of the homes. The negative numbers in Table 4.8 identify a decrease of UFP concentration indoor due to the penetration of air from ventilation. The highest decrease of UFP indoors was of 2.02×10^4 particle cm^{-3} and the highest increase in indoor UFP concentration was 7.45×10^2 particle cm^{-3}. Both these values were observed in H2.

The use of electric oven was the activity that resulted in the overall highest increase of UFP indoor concentrations, being followed by the usage of electric toaster and boiling in cooker. Smoking resulted in lower increases of UFP in comparisons with these three activities but had a cumulative influence on UFP indoor concentrations, lingering these high levels for a longer period of time. Smoking also seems to have a determinant effect of the formation of PM as we can observe further in section 4.3.1.

In literature, we can find some studies that measured the peak concentration produced by specific indoor sources in controlled environment. The existing studies are described in the following Table 4.3.2.

Table 4.3.2 - Chamber studies relating UFP producing activities indoors and associated levels.

Source	Particle	Levels	Study Conditions	Reference
Vacuuming	UFP	$0.04 \pm 0.02 \times 10^{11}$ particles min^{-1}		(Wu et al., 2011)
		2.14×10^{4} particles cm^{-3}	With full bag	(Afshari et al., 2005)
		3.83×10^{4} particles cm^{-3}	Motor without bag	
Ironing	UFP	5.50×10^{2} particles cm^{-3}	Without steam	(Afshari et al., 2005)
		7.2×10^{3} particles cm^{-3}	With steam	
Cigarette smouldering		$36 \pm 0.24 \times 10^{11}$ particles min^{-1}		(Wu et al., 2011)
		2.13×10^{5} particles cm^{-3}		(Afshari et al., 2005)
Candle burning	UFP	$4.92 \pm 3.23 \times 10^{4}$ particles cm^{-3}		(Glytsos et al., 2010)
		2.41×10^{5} particles cm^{-3}	Pure wax	(Afshari et al., 2005)
		6.96×10^{4} particles cm^{-3}	Scented candles	
Incense Burning	UFP	$0.44 \pm 0.33 \times 10^{11}$ particles min^{-1}		(Wu et al., 2011)
Cooking: Gas combustion	UFP	$(2.59 \pm 0.89) \times 10^{11}$ particles min^{-1}		(Wu et al., 2011)
		7.9×10^{4} particles cm^{-3}	Propane gas cooker	(Afshari et al., 2005)

		UFP	2.6 ×10^4 particles cm^{-3}	1 heating ring	(Dennekamp et al., 2001)
			1.46 × 10^5 particles cm^{-3}	4 heating rings	
	Electric cooker	UFP	1.11 × 10^5 particles cm^{-3}		(Afshari et al., 2005)
			9.4 ×10^4 particles cm^{-3}	1 heating ring	(Dennekamp et al., 2001)
			1.11 × 10^5 particles cm^{-3}	4 heating rings	
	Oven	Bake cake	UFP	9.8 × 10^4 particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
				3.0 × 10^4 particles cm^{-3}	Electric fuel	
		Roast meat	UFP	1.24 × 10^5 particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
				2.4 × 10^4 particles cm^{-3}	Electric fuel	
		Bake Potatoes	UFP	1.25 × 10^5 particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
				1.6 ×10^4 particles cm^{-3}	Electric fuel	
	Grilling	Only grilling	UFP	1.03 × 10^5 particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
				7.7 × 10^4 particles cm^{-3}	Electric fuel	
		Toast	UFP	1.38 × 10^5 particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
				1.34 X 10^5 particles cm^{-3}	Electric fuel	
		Grill bacon	UFP	4.13 ×10^5 particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
				5.30 ×10^5 particles cm^{-3}	Electric fuel;	
	Heating an empty pan		UFP	(58.36 ± 40.60) × 10^{11} particles min^{-1}		(Wu et al., 2011)

	Boiling	UFP	$5.33 \pm 3.89 \times 10^{11}$ particles min^{-1}		(Wu et al., 2011)
			1.33×10^{5} particles cm^{-3}		(Dennekamp et al., 2001)
	Steaming	UFP	$4.70 \pm .57 \times 10^{11}$ particles min^{-1}	Gas fuel	(Wu et al., 2011)
	Pan frying	UFP	$104.40 \pm 31.44 \times 10^{11}$ particles min^{-1}		(Wu et al., 2011)
			1.5×10^{5} particles cm^{-3}		(Afshari et al., 2005)
	Stir frying	UFP	$148.29 \pm 46.60 \times 10^{11}$ particles min^{-1}	Meet frying	(Wu et al., 2011)
			1.37×10^{5} particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
			1.1×10^{4} particles cm^{-3}	Electric fuel	
	Fry bacon	UFP	5.9×10^{5} particles cm^{-3}	Gas fuel	(Dennekamp et al., 2001)
			1.5×10^{5} particles cm^{-3}	Electric fuel	

Comparing the obtained levels of UFP in four homes with those of the chamber studies it was possible to conclude that ironing corresponded with the expected values (ironing with steam). Cigarette smouldering was reported to have slightly higher levels of UFP than obtained in this study. UFP levels reported due to boiling corresponded to the range of increases obtained in all the homes. Frying also resulted in increases in UFP levels in homes similar to the concentration increases of the chamber studies.

The following figures illustrate the increases of UFP levels due to some specific activities observed in four homes. The examples were selected from sampling at H1, H2, H3 and H4 and are present in the following Figures 4.3.1 - 4.3.4.

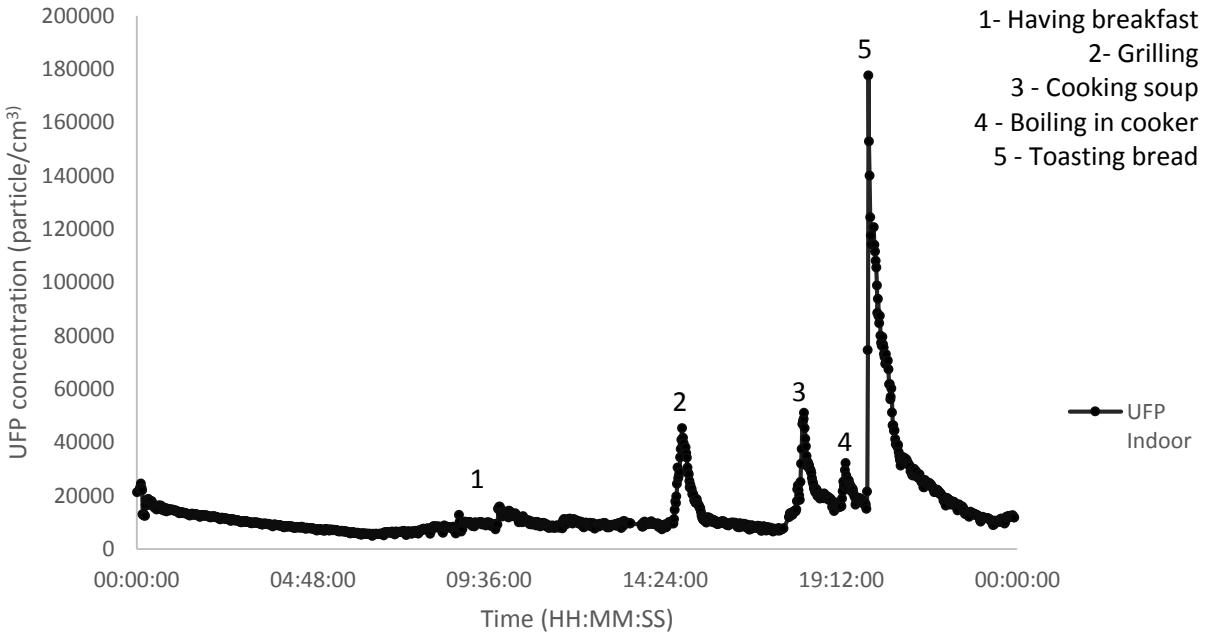


Figure 4.3.1 - Example of activities impact on UFP concentration in H1 (09.04.2014).

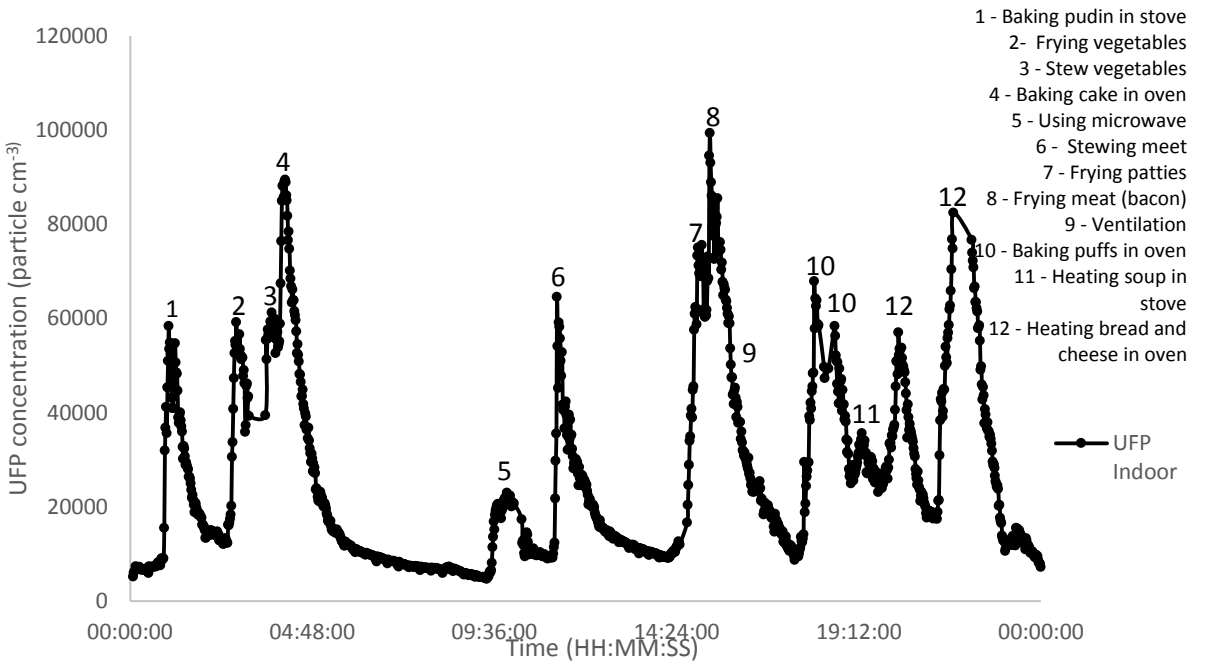


Figure 4.3.2 - Example of activities impact on UFP concentration in H2 (20.04.2014).

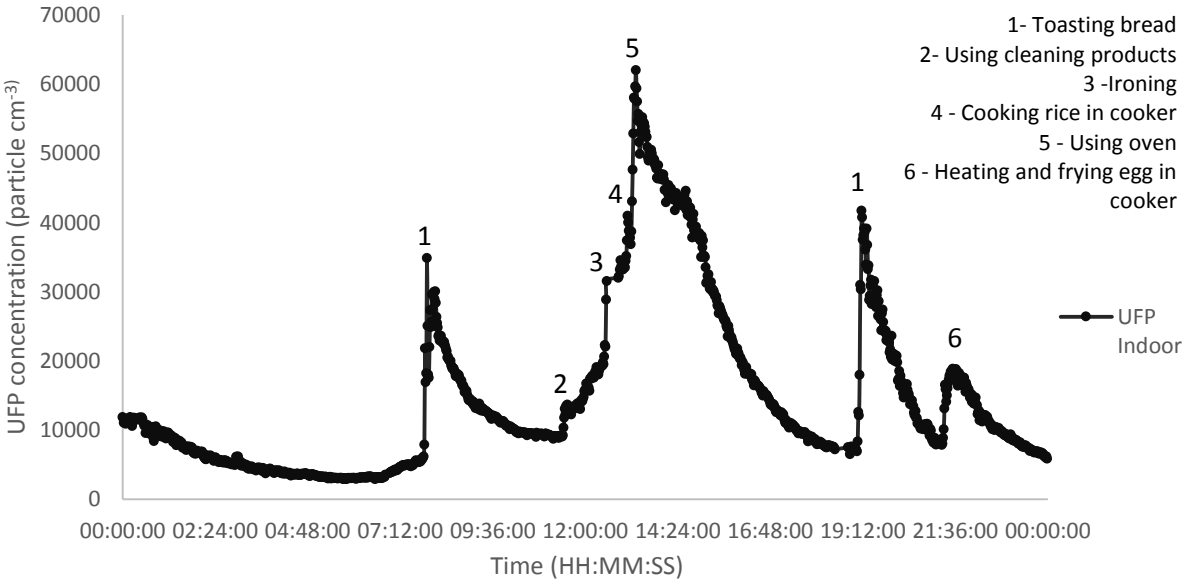


Figure 4.3.3 - Example of activities impact on UFP concentration in H3 (07.05.2014).

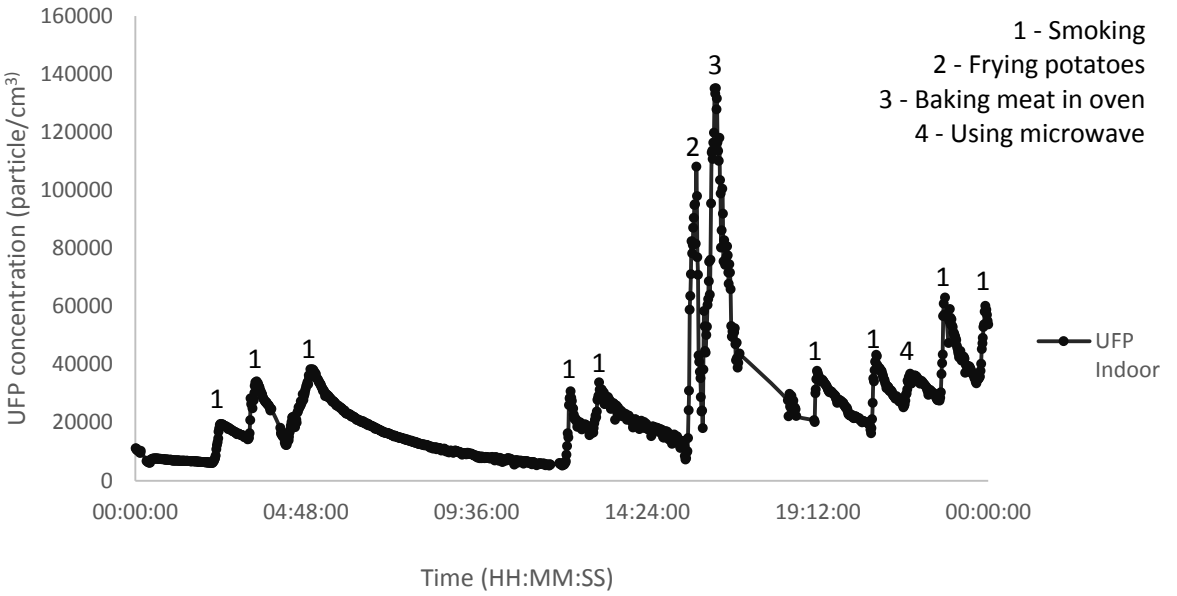


Figure 4.3.4- Example of activities impact on UFP concentration in H4 (21.05.2014).

4.3.1 Influence of UFP formation in PM

Sources of UFP have shown to be also associated with and consequently to influence PM levels indoors, especially smoking. Figure 4.3.5 illustrates a daily profile of UFP and PM levels in the smoking home.

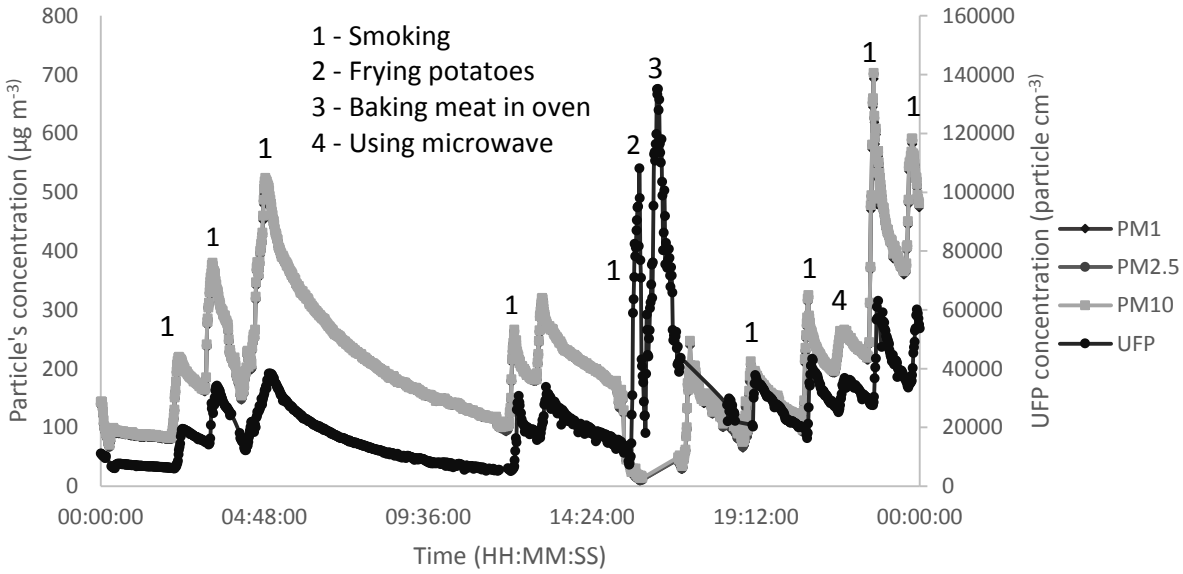


Figure 4.3.5 - Example UFP and PM in H4 (21.05.2014).

As can be clearly observed, there is an obvious correlation between UFP and PM daily profile, especially when the producing activity is smoking. Frying and baking meet do not seem to have so strong influence on PM levels.

4.4 Assessment of Exposure Doses

Age specific dose rates were calculated in order to evaluate the potential exposure to UFP in home environment. An analysis of daily activities inside the homes was made with the resource of a specific questionnaire for this purpose. Total daily residence time of children and adults in homes and the intensity of their activities was registered in order to obtain corresponding breathing rates. Table 4.4.1 and Table 4.4.2 show the parameters used from exposure dose rate calculation at four homes.

Table 4.4.1- Main parameters for the calculation of dose rates to 3 to 5 years old children.

Parameter	***H1***	***H2***	***H3***	***H4***
	3 to 5 years old	**3 to 5 years old**	**3 to 5 years old**	**3 to 5 years old**
BR_{WA} (l min^{-1})	5.88	5.88	5.88	5.88
C_{WA} (particle number cm^{-3})	1.24×10^4	1.09×10^4	1.11×10^4	1.62×10^4
N (h)	13.00	13.0	13.0	13.0
BW (kg)	18.6	18.6	18.6	18.6

Table 4.4.2 - Main parameters for the calculation of dose rates to 21 to 60 years old adults.

Parameter	***H1***	***H2***	***H3***	***H4***
	21 to 60 years old	**21 to 601 years old**	**21 to 60 years old**	**21 to 60 years old**
BR_{WA} (l min^{-1})	6.53	6.53	6.53	6.53
C_{WA} (particle number cm^{-3})	1.24×10^4	1.09×10^4	1.11×10^4	1.62×10^4
N (h)	13.00	13.0	13.0	13.0
BW (kg)	76	76	76	76

The calculated does rates of UFP for the occupants in the four homes are presented in Table 4.4.2 as well as the comparison with a does rates estimated in a previous study on indoor UFP in schools for children (Fonseca, 2013).

Table 4.4.3 - Calculated age-specific dose rates of UFP for children in for four homes: a comparison with school environments.

	Homes				
	H1	***H2***	***H3***	***H4***	***Reference***
Dose (particle number $kg^{-1}d^{-1}$)	**3 to 5 years old**	**3 to 5 years old**	**3 to 5 years old**	**3 to 5 years old**	This study
	3.04×10^{3}	2.69×10^{3}	2.73×10^{3}	4.00×10^{3}	
	21 to 60 years old	**21 to 60 years old**	**21 to 60 years old**	**21 to 60 years old**	
	8.27×10^{2}	7.32×10^{2}	7.43×10^{2}	1.09×10^{3}	
	Schools				
	US1		***US2***	***RS1***	***Reference***
	3 to 5 years old		**3 to 5 years old**	**3 to 5 years old**	(Fonseca, 2013)
	4.60×10^{9}		1.84×10^{9}	4.50×10^{9}	

Overall, it is possible to conclude that the highest dose rates of UFP was observed in the homes with higher indoor UFP levels (H4). Comparing the results obtained for children with a recent assessment on indoor UFP in schools, it was possible to observe that the calculated dose rates for 3-5 years old children in home environments are much lower ($4.6\times10^{5} – 1.7\times10^{5}$) than in schools. It is somehow surprising considering the longer amount of time that children spend at home environments and overall lower UFP levels reported for the schools by Fonseca (2013). The main differences were the type of activity performed by children in these two different environments. Contrary to schools, most of the time children spent at home was performing low intensity activities, such as sleeping or sedentary activities, during the day (playing, TV watching, reading) which imply much lower breathing rates. This translates in significantly lower dose rates of UFP in the homes, although the overall indoor UFP concentration in homes were generally higher than in schools. These results demonstrate that in order to correctly assess child overall exposure it is necessary to always consider not only the levels of pollution of indoor specific places but the activities performed. Comparing adults dose rates with children dose rates, we can observe they are significantly lower ($1.8\times10^{-1} – 4.1\times10^{-1}$) due to the much higher body weight of adults. Although they are in contact with the same particle number concentration as present higher breathing rates, children's lower body weight is a determinant factor in the resulting exposure dose. This underlines the importance and susceptibility of the special group of children regarding exposure to indoor UFP.

4.5 References

AFSHARI, A., MATSON, U. & EKBERG, L. E. 2005. Characterization of indoor sources of fine and ultrafine particles: a study conducted in a full-scale chamber. *Indoor air,* 15**,** 141-150.

BEGUM, B. A., PAUL, S. K., HOSSAIN, M. D., BISWAS, S. K. & HOPKE, P. K. 2009. Indoor air pollution from particulate matter emissions in different households in rural areas of Bangladesh. *Building and Environment,* 44**,** 898-903.

BEKÖ, G., WESCHLER, C. J., WIERZBICKA, A., KAROTTKI, D. G., TOFTUM, J., LOFT, S. & CLAUSEN, G. 2013. Ultrafine particles: Exposure and source apportionment in 56 Danish homes. *Environmental Science and Technology,* 47**,** 10240-10248.

BHANGAR, S., MULLEN, N., HERING, S., KREISBERG, N. & NAZAROFF, W. 2011. Ultrafine particle concentrations and exposures in seven residences in northern California. *Indoor Air,* 21**,** 132-144.

CATTANEO, A., PERUZZO, C., GARRAMONE, G., URSO, P., RUGGERI, R., CARRER, P. & CAVALLO, D. M. 2011. Airborne particulate matter and gaseous air pollutants in residential structures in Lodi province, Italy. *Indoor Air,* 21**,** 489-500.

CHAO, C. Y. & WONG, K. K. 2002. Residential indoor PM10 and PM2.5 in Hong Kong and the elemental composition. *Atmospheric Environment,* 36**,** 265-277.

DENNEKAMP, M., HOWARTH, S., DICK, C., CHERRIE, J., DONALDSON, K. & SEATON, A. 2001. Ultrafine particles and nitrogen oxides generated by gas and electric cooking. *Occupational and Environmental Medicine,* 58**,** 511-516.

DIAPOULI, E., ELEFTHERIADIS, K., KARANASIOU, A. A., VRATOLIS, S., HERMANSEN, O., COLBECK, I. & LAZARIDIS, M. 2011. Indoor and outdoor particle number and mass concentrations in Athens. Sources, sinks and variability of aerosol parameters. *Aerosol and Air Quality Resarch,* 11**,** 632-642.

FITTSCHEN, U. E. A., SANTEN, M., REHMERS, A., DURUKAN, I. & WESSELMANN, M. 2013. Indoor Aerosol Determination with Respect to a Soiling Phenomenon in Private Residences. *Environmental Science & Technology,* 47**,** 608-615.

FONSECA, J. A. C. 2013. *Indoor Ultrafine Particles: Evaluation of Pre-School Environments.* Integrated Master in Environmental Engineering, Faculdade de Engenharia da Universidade do Porto.

GLYTSOS, T., ONDRÁČEK, J., DŽUMBOVÁ, L., KOPANAKIS, I. & LAZARIDIS, M. 2010. Characterization of particulate matter concentrations during controlled indoor activities. *Atmospheric Environment,* 44**,** 1539-1549.

HÉROUX, M.-E., CLARK, N., RYSWYK, K. V., MALLICK, R., GILBERT, N. L., HARRISON, I., RISPLER, K., WANG, D., ANASTASSOPOULOS, A. & GUAY, M. 2010. Predictors of indoor air concentrations in smoking and non-smoking residences. *International journal of environmental research and public health,* 7**,** 3080-3099.

KEARNEY, J., WALLACE, L., MACNEILL, M., HÉROUX, M.-E., KINDZIERSKI, W. & WHEELER, A. 2014. Residential infiltration of fine and ultrafine particles in Edmonton. *Atmospheric Environment,* 94**,** 793-805.

KEARNEY, J., WALLACE, L., MACNEILL, M., XU, X., VANRYSWYK, K., YOU, H., KULKA, R. & WHEELER, A. 2011. Residential indoor and outdoor ultrafine particles in Windsor, Ontario. *Atmospheric Environment,* 45**,** 7583-7593.

LEAVEY, A., PEREZ, H. R. & BURSTYN, I. 2012. Endotoxin and Particle Concentrations in Subsidized Households with Asthmatic Children in Philadelphia: A Pilot Study. *Indoor and Built Environment,* 21**,** 515-523.

MCAULEY, T., FISHER, R., ZHOU, X., JAQUES, P. & FERRO, A. 2010. Relationships of outdoor and indoor ultrafine particles at residences downwind of a major international border crossing in Buffalo, NY. *Indoor air,* 20**,** 298-308.

MCCORMACK, M. C., BREYSSE, P. N., MATSUI, E. C., HANSEL, N. N., WILLIAMS, D., CURTIN-BROSNAN, J., EGGLESTON, P. & DIETTE, G. B. 2009. In-home particle concentrations and childhood asthma morbidity. *Environ Health Perspect,* 117**,** 294-298.

MORAWSKA, L., HE, C., HITCHINS, J., MENGERSEN, K. & GILBERT, D. 2003. Characteristics of particle number and mass concentrations in residential houses in Brisbane, Australia. *Atmospheric Environment,* 37**,** 4195-4203.

MULLEN, N. A., LIU, C., ZHANG, Y., WANG, S. & NAZAROFF, W. W. 2011. Ultrafine particle concentrations and exposures in four high-rise Beijing apartments. *Atmospheric Environment,* 45**,** 7574-7582.

OGULEI, D., HOPKE, P. & WALLACE, L. 2006. Analysis of indoor particle size distributions in an occupied townhouse using positive matrix factorization. *Indoor Air,* 16**,** 204-215.

PRASAUSKAS, T., MARTUZEVICIUS, D., KRUGLY, E., CIUZAS, D., STASIULAITIENE, I., SIDARAVICIUTE, R., KAUNELIENE, V., SEDUIKYTE, L., JURELIONIS, A. & HAVERINEN-SHAUGHNESSY, U. 2014. Spatial and temporal variations of particulate matter concentrations in multifamily apartment buildings. *Building and Environment,* 76**,** 10-17.

WALLACE, L. & HOWARD-REED, C. 2002. Continuous monitoring of ultrafine, fine, and coarse particles in a residence for 18 months in 1999-2000. *Journal of the Air & Waste Management Association,* 52**,** 828-844.

WHEELER, A. J., WALLACE, L. A., KEARNEY, J., VAN RYSWYK, K., YOU, H., KULKA, R., BROOK, J. R. & XU, X. 2011. Personal, Indoor, and Outdoor Concentrations of Fine and Ultrafine Particles Using Continuous Monitors in Multiple Residences. *Aerosol Science and Technology,* 45**,** 1078-1089.

WU, C., CHAO, C. Y., SZE-TO, G., WAN, M. & CHAN, T. 2011. Ultrafine particle emissions from cigarette smouldering, incense burning, vacuum cleaner motor operation and cooking. *Indoor and Built Environment***,** 1420326X11421356.

5. Conclusion and Future Perspectives

5.1 Conclusions

The execution of this study had led to the following conclusions.

The indoor UFP concentrations in residences are fully dependent on the existent sources and living habits of the occupants. This said, it is determinant if smoking is performed indoors, since it is a major cumulative source of air pollutants, namely of PM and UFP. Type of cooking activities and the number of meals cooked in the home also greatly influenced UFP number concentration in homes. Another factor that showed to have a great influence on the levels of particles is room volume. The higher number concentrations emitted from the same source were found in spaces with smaller volume. The decrease of UFP number concentration took longer if the number of room occupants is higher in relation to room volume. Cooking with the closed doors and ventilation the kitchen after the activities like the usage of oven or electric toaster may be advisable in order to achieve healthier air in homes.

Higher indoor PM values were also found in the home with smokers, allowing to conclude that tobacco smoke is one of the major contributor for indoor PM.

Regarding outdoor UFP number concentration, the lower levels were observed in a rural area and sub-urban areas.

Indoor/outdoor ratio for UFP particles indicated that indoor sources were the major contributors for indoor UFP levels. I/O of $PM_{2.5}$ were also higher than 1 for all homes, leading to the conclusion that indoor sources were the major contributor for this fraction mass concentration indoors.

The levels of indoor particles obtained in this study were in general agreement to other international studies.

The obtained exposure dose rates of UFP for 3 to 5 years old children were significantly lower in homes than in school environments, mostly due to the type and intensity of activities performed indoors. 21 to 60 years old dose rates of UFP in homes were much lower than children's, as expected due to their higher body weight.

In addition, indoor concentrations of UFP particles varied more greatly in homes than in schools, because there is vast number of indoor sources inside the homes that do not exist in schools.

5.2 Future Perspectives

In order to improve this work, higher number of homes would benefit the results obtained. Also, homes with more similar characteristics between them would be advisable for a better interpretation of the obtained results.

Sampling collection in different microenvironments inside each home would be useful to assess more accurately dose rate to UFP well as to better understand particle's dynamics inside the confined spaces.

The repetition of the sampling campaign during different seasons would be also useful to this work, since UPF number concentrations outdoors, and consequently indoors, vary significantly between seasons.

APPENDIX

A. Home Characterization

In order to better understand the conditions of the sampling campaign and how the existing characteristics of each building and its surroundings affected the results obtained in this study, the following characterization aims to correlate possible influential factors.

A.1 Home 1

The following micro-environments properties' for H1 are presented in this sub-chapter.

A.1.1 Living/Dining Room

Table 1 - Relevant H1 Living/Dining room properties.

Total Room Volume (m^3)	66.05
Total Room Surface (m^2)	109.14
Ventilation (Natural / Forced)	Natural
Heating Systems (Existent / Non-existent)	Non-existent
Room Height (m)	2.703
Main Materials	Wood, Cotton, Wall
Floor Coating	Wood
Wall Coating	White Paint
Ceiling Coating	White Paint
Door Surface (m^2)	1.58
Type of Door	Wood
Number of Doors	2
Window Surface (m^2)	2.889
Type of Window	Double glass layer and aluminium
Number of Windows	2
Average Temperature (˚C)	23.43
Average Relative Air Humidity (%)	53.28

Daily Sampling Period	00:00 to 23:59
Sampling Dates	08.04.2014 to 16.04.2014
Distance from sampling point to main exit (m)	2.718

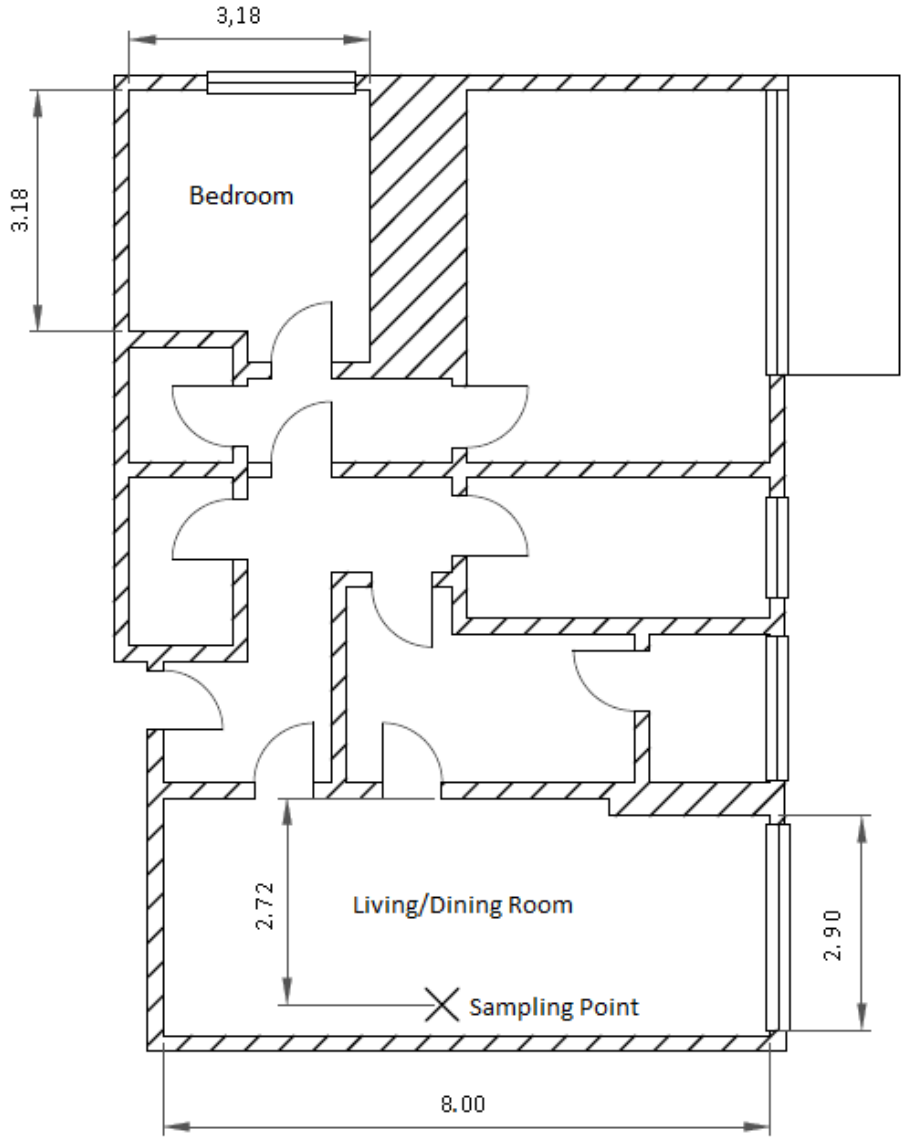


Figure 1 - H1 estimated blueprint.

Figure 2 – H1 Indoor sampling.

Figure 3 - H1 Indoor sampling site - Living/Dining Room (1).

Figure 4 - H1 Indoor sampling site - Living/Dining Room (2).

Figure 5 - H1 Outdoor sampling site - Bedroom window.

A.2 Home 2

A.1.2 Living/Dining Room

Table 2 - Relevant H2 Living/Dining room properties.

Total Room Volume (m^3)	117.28
Total Room Surface (m^2)	171.08
Ventilation (Natural / Forced)	Natural
Heating Systems (Existent / Non-existent)	Existing – Fireplace (never used during campaign)
Room Height (m)	2.90
Main Materials	Wood, Wall, Tile, Cotton
Floor Coating	Tile
Wall Coating	Paint
Ceiling Coating	Paint
Door Surface (m^2)	2.40
Type of Door	Wood and glass
Number of Doors	3
Window Surface (m^2)	5.00
Type of Window	Double glass layer and aluminium
Number of Windows	2
Average Temperature (°C)	21.70
Average Relative Air Humidity (%)	54.70
Daily Sampling Period	00:00 to 23:59
Sampling Dates	16.04.2014 to 22.04.2014; 24.04.2014 to 28.04.2014
Distance from sampling point to main exit (m)	3.50

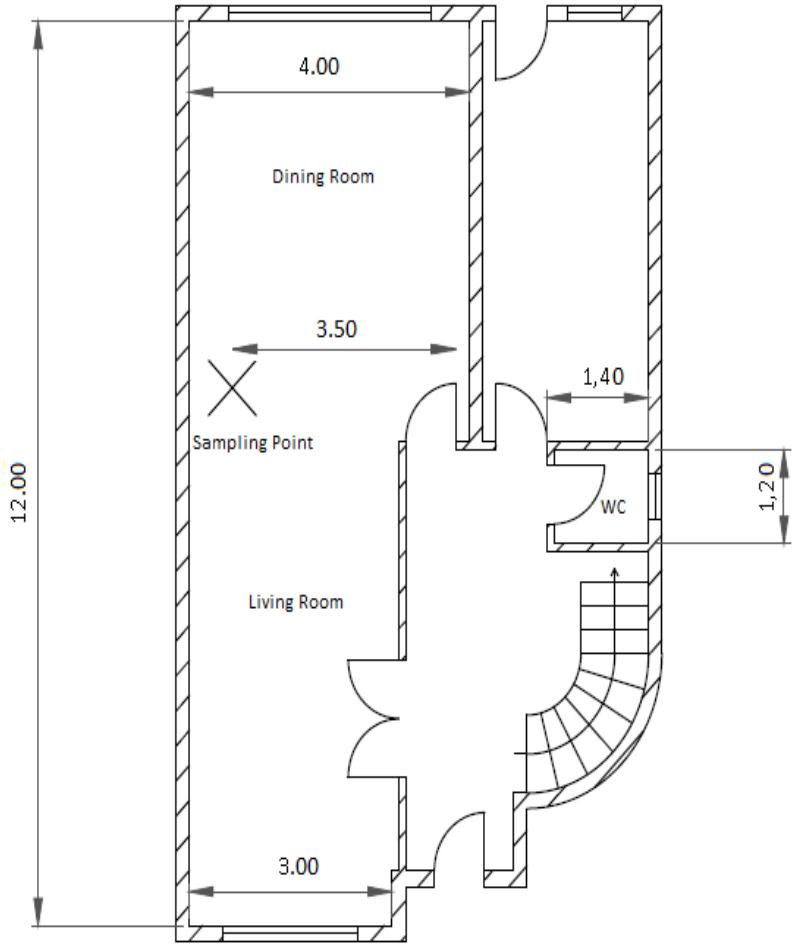


Figure 6 - H2 blueprint.

Figure 7 – H2 Indoor sampling site –Living/Dining room (1)

Figure 5.28 – H2 Indoor sampling site –Living/Dining room (2)

Figure 9 - H2 Outdoor sampling site - WC window

A.3 Home 3

A.1.3 Living/Dining Room

Table 3 - Relevant H3 Living/Dining room properties.

Total Room Volume (m^3)	116.10
Total Room Surface (m^2)	-
Ventilation (Natural / Forced)	Natural
Heating Systems (Existent / Non-existent)	Existing – Fireplace (never used during campaign)
Room Height (m)	2.70
Main Materials	Wood, Wall, Cotton
Floor Coating	
Wall Coating	Pait
Ceiling Coating	Paint
Door Surface (m^2)	-
Type of Door	Wood
Number of Doors	1
Window Surface (m^2)	-
Type of Window	Double glass layer and aluminium
Number of Windows	2
Average Temperature (˚C)	22.35
Average Relative Air Humidity (%)	60.40
Daily Sampling Period	00:00 to 23:59
Sampling Dates	02.05.2014 to 10.05.2014
Distance from sampling point to main exit (m)	-

"-": Not available

A.4 Home 4

A.1.4 Living/Dining Room

Table 4 - Relevant H4 Living/Dining room properties.

Total Room Volume (m^3)	158.63
Total Room Surface (m^2)	203.38
Ventilation (Natural / Forced)	Natural and Forced (AVAC system)
Heating Systems (Existent / Non-existent)	Existent (AVAC system)
Room Height (m)	2.70
Main Materials	Wood, Wall, Paint, Cotton
Floor Coating	Wood
Wall Coating	Paint
Ceiling Coating	Paint
Door Surface (m^2)	2.60
Type of Door	Wood
Number of Doors	2
Window Surface (m^2)	10.13 and 14.90
Type of Window	Double glass layer and aluminium
Number of Windows	2
Average Temperature (˚C)	21.62
Average Relative Air Humidity (%)	59.17
Daily Sampling Period	00:00 to 23:59
Sampling Dates	15.04.2014 to 22.04.2014
Distance from sampling point to main exit (m)	-

"-": Not available

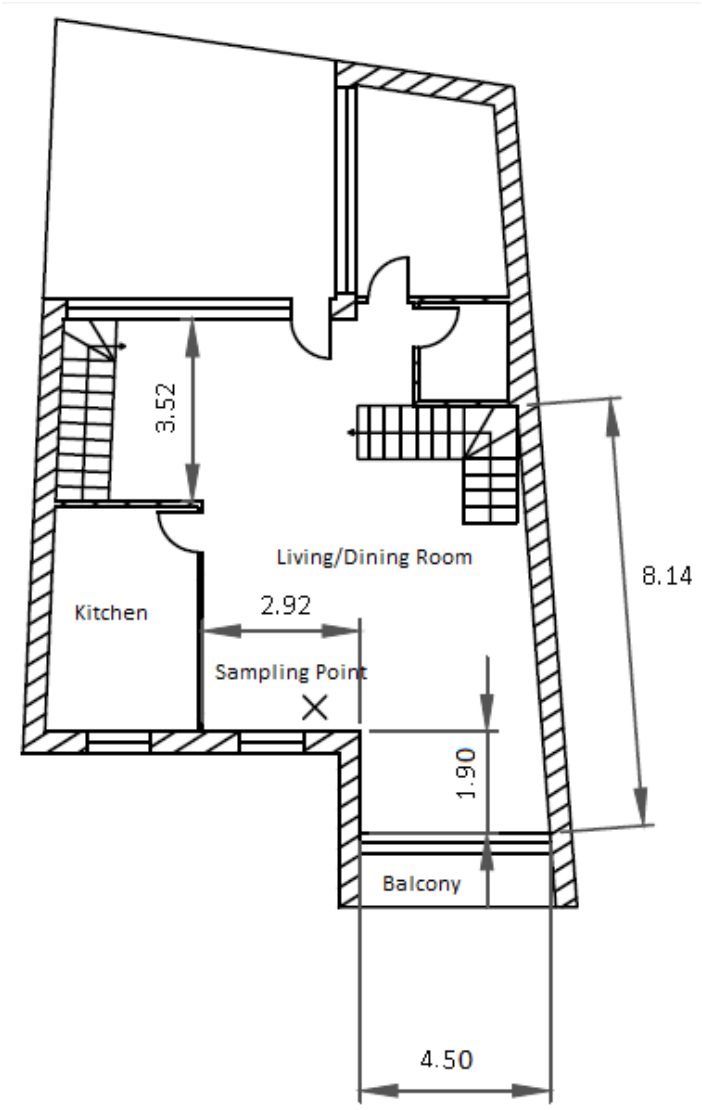


Figure 10- H4 blueprint.

Figure 11 - H4 Indoor sampling.

Figure 12 - H4 Indoor sampling site - Living/Dining room (1).

Figure 13 - H4 Indoor sampling site - Living/Dining room (2).

Figure 14 - Outdoor sampling site - Living room balcony.

B. Questionnaires

In order to better understand the conditions of the sampling campaign and the possible factors influencing study, the occupants were asked to fill daily the following questionnaires.

B.1 Room occupancy

The occupancy of the room where the sampling took place as well as the respective time period were daily registered in a questionnaire present in the following Table 5.

Table 5 - Room occupancy questionnaire.

Room Occupancy
Date:

00h	
01h	
02h	
03h	
04h	
05h	
06h	
07h	
08h	
09h	
10h	
11h	
12h	
13h	
14h	
15h	
16h	
17h	
18h	
19h	
20h	
21h	
22h	
23h	

B.2 Indoor Sources

Potential sources of UFP and PM and the respective period of time they occurred were daily registered in a questionnaire, present bellow in Table 6.

Table 6 – UFP and PM sources registry questionnaire.

Date:

Ultrafine Particles		
Cooking	Soup cooking	
	Boiling	
	Toasting	
	Frying	
	Baking	
	Grilling	
Use of electric toaster		
Use of electric cooker		
Candle or incense burning		
Smoking		
Use of printers (-ink)		
Hair spraying		
Use of cleaning products		
Use of furniture polish		
Ironing		
Ventilation		
Particulate Matter		
Human movements		
Sitting on the furniture		
Folding of clothes		
Hovering		
Dusting		
Sweeping		
Other		

B.3 Children's Activities

In order to assess exposure dose for children living in the sampled residences, their activities while at home and the period they occurred were daily registered in the following Table7.

Table 7 - Children's activities questionnaire.

Children's Activities
Date:

08h	
09h	
10h	
11h	
12h	
13h	
14h	
15h	
16h	
17h	
18h	
19h	
20h	
21h	
22h	
23h	